# Far-infrared study of free carriers and the plasmon-phonon interaction in CdTe†

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The reflectivity of $n$-CdTe has been measured between 16 and 203 $cm^{-1}$ at room temperature. The single-crystal samples had carrier concentrations $N$ between $5\times10^{16}$ and $1\times10^{18}$ $cm^{-3}$. The data could be fitted by using classical lattice theory and Drude-type free-carrier theory with an energy-independent scattering time. The values of $N$ and mobility $\mu$ obtained from the fits agreed reasonably well with Hall and resistivity values. The agreement was not improved by the introduction of an energy-dependent scattering time for either polar-mode or ionized-impurity scattering. The plasmon-phonon minima were observed and their frequencies were found to be strongly influenced by $\mu$ as well as by $N$. The optical lattice parameters were determined as follows: $\epsilon_s = 10.2$, $\epsilon_\infty = 7.1$, $\omega_t = 141$ $cm^{-1}$, and $\Gamma = 8.7$ $cm^{-1}$. These results agree well with previous measurements.

## INTRODUCTION

The examination of a semiconductor in the far infrared can be of interest for at least two reasons. First, as previous work has shown, [1] the optical data can be easily related to the semiconductor transport properties because at these long wavelengths a simple non-quantum-mechanical optical theory holds. Second, over a wide range of carrier concentration in most semiconductors, the lower branch of the coupled plasmon-phonon mode falls almost wholly in the far infrared and its frequency can be observed at the reflectivity minimum. In this paper we report on measurements of far-infrared reflectivity in $n$-CdTe with results that relate to both the transport properties and the plasmon-phonon coupling. The plasmon-phonon behavior is of particular interest in CdTe since, unlike many other polar semiconductors, its transverse-optical frequency lies in the far infrared.

CdTe has previously been studied in the far infrared[2-8] as well as by Raman scattering. [9] The plasmon-phonon interaction was observed in the Raman experiment but was not examined as a function of free-carrier concentration and scattering time. Similarly, although the transport behavior was touched on in some of the far-infrared work, no detailed study has been made over a range of carrier concentration and the appearance of the plasmon-phonon minimum in the reflectivity has not been analyzed. In this work we examine four samples with carrier concentrations ranging from $5\times10^{16}$ to $1\times10^{18}$ $cm^{-3}$. For these concentrations at room temperature, where our measurements were carried out, the carriers are in the classical-to-degenerate transition region. The connection between the optical data and the transport properties is complicated by this partial degeneracy and by the fact that several scattering mechanisms, especially polar-optical-mode and ionized-impurity scattering, contribute to the mobility. [10] We analyze our data with an extensive use of scattering theory and carry out a detailed analysis of the plasmon-phonon minimum as well.

## EXPERIMENTAL DETAILS

Our samples were grown and characterized at the M. I. T. Lincoln Laboratory. The sample properties are given in Table I. The characterization was made by means of low-field bar-geometry Hall and resistivity measurements. The values shown in the table were calculated assuming a Hall scat-

TABLE I. Comparison of electrical and optical carrier parameters. No adjustment has been made for energy-dependent electron scattering. The errors quoted for the optical numbers are standard deviations obtained from the least-squares-fit program.[a] The experimental errors associated with the electrical measurements are on the order of 10%.[b]

| Sample | Dopant | Carrier concentration ($10^{18}$ $cm^{-3}$) Electrical | Optical | Mobility ($cm^2$/V sec) Electrical | Optical | Resistivity (Ω cm) Electrical | Optical |
|---|---|---|---|---|---|---|---|
| 9-71/2 | Ga | 0.051 | 0.13±0.04 | 410 | 200±60 | 0.30 | 0.24±0.15 |
| 5-71/E | Ga | 0.14 | 0.15±0.01 | 690 | 480±30 | 0.067 | 0.087±0.01 |
| 32-70/I | I | 0.41 | 0.34±0.02 | 910 | 700±60 | 0.017 | 0.026±0.004 |
| 4-70/A7 | Al | 1.3 | 1.1±0.1 | 760 | 760±80 | 0.0063 | 0.0075±0.002 |

[a]Reference 14.
[b]A. J. Strauss (private communication).

tering factor of unity. The samples were supplied with mechanically polished surfaces.

Three of the CdTe samples as received were not of convenient size and shape for the reflectivity measurements. Plates about 2×4×4 mm were cut from each of these samples. One surface of each plate was left as received and the opposite surface was lapped and polished. The final polish was done with 0.3-$\mu$m aluminum oxide micropolish. In one case the polished surface was later subjected to a bromine etch. For all the samples, reflection spectra were taken on the surface as received, on the lapped and polished surface, and on the bromine-etched surface. In only one sample, 32-70/I, was there any measurable difference between the reflectivity of the surface as received and the reflectivity of the newly polished surface. After removal of about 10 $\mu$m from the as-received surface the two spectra agreed and these spectra were taken as representative of the bulk material.

Reflectivity measurements were made by the Fourier-transform method using a commercial Grubb-Parsons Michelson interferometer with a Unicam Golay detector. The instrument was modified so that reflectivity measurements could be made at a 12° angle of incidence. Reflectivity at this angle of incidence differs negligibly from that at normal incidence. The sample-in–sample-out technique was used with the reflection from either a polished coin-silver or stainless-steel mirror providing the reference. In the case of the stainless-steel reference the measured reflection was adjusted according to standard optical theory because of the relatively high resistivity of stainless steel. Calculations show that the reflectivity of coin silver should differ negligibly from unity. Data were taken from 16 to 203 $cm^{-1}$ with a resolution of 8 $cm^{-1}$. Mylar beamsplitters of 6.25-, 12.5-, and 25-$\mu$m thickness were used in order to improve the data over various frequency intervals. For each sample, several sets of data were averaged together to provide an improved signal-to-noise ratio and a measure of the uncertainty in the spectrum.

## THEORY AND DATA ANALYSIS

The reflection coefficient for a semi-infinite medium in the case of normal incidence is given by

$$R = [(n-1)^2 + k^2]/[(n+1)^2 + k^2], \tag{1}$$

where $n$ and $k$ are the real and imaginary parts of the complex index of refraction $\tilde{n}$ and

$$\tilde{n}^2 = \epsilon(\omega), \tag{2}$$

where $\epsilon$ is the complex dielectric function and $\omega$ is the optical frequency. The dielectric function can be written as the sum of a lattice term and a free-carrier term

$$\epsilon = \epsilon_{\mathrm{lat}} + \epsilon_{fc}. \tag{3}$$

The lattice contribution is given by[11]

$$\epsilon_{\mathrm{lat}} = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 - (\omega/\omega_t)^2 + i\omega\Gamma/\omega_t^2}, \tag{4}$$

where $\epsilon_s$ and $\epsilon_\infty$ are the low- and high-frequency dielectric constants. The lattice dielectric function exhibits a resonance at the transverse-optical frequency $\omega_t$, with a damping term given by $\Gamma$. The values of the four lattice parameters in CdTe have been measured by several workers using different techniques. The means of the values reported in the literature, together with the variation from measurement to measurement expressed as a standard deviation, are given in Table II.

The free-carrier term is given by[12]

$$\epsilon_{fc}(\omega) = \omega_p^2 \epsilon_\infty \left( -\left\langle \frac{\tau_k^2}{1+\omega^2\tau_k^2} \right\rangle + \frac{i}{\omega} \left\langle \frac{\tau_k}{1+\omega^2\tau_k^2} \right\rangle \right), \tag{5}$$

where the plasma frequency $\omega_p$ is defined by

$$\omega_p^2 = 4\pi N e^2/m^* \epsilon_\infty. \tag{6}$$

Here, $N$ is the free-electron concentration, $e$ is the electronic charge, $m^*$ is the electronic effective mass, and $\tau_k$ is the energy-dependent electron scattering time. The angular brackets indicate averaging over the electron-distribution function. The effective mass is taken as $0.11 m_0$ as determined by Marple.[13] No polaron correction to the effective mass was included in the optical theory. The electron scattering time is related to the drift mobility $\mu$ by the equation

$$\mu = e\langle \tau_k \rangle / m^*. \tag{7}$$

The reflectivity data were fitted with a simplified version of the theory presented above. Because of the complexity involved in the averaging of the functions of $\tau_k$ appearing in Eq. (5), the reflectivity calculations were carried out assuming

TABLE II. Comparison of the mean values of CdTe lattice parameters taken from the literature[a] with those determined from least-squares fits to the reflectivity data reported in this paper. The errors for the literature values are the standard deviations of the reported numbers. The errors reported for this work are standard deviations as determined from the least-squares fits to the data.

| | $\epsilon_s$ | $\epsilon_\infty$ | $\omega_t$($cm^{-1}$) | $\Gamma$($cm^{-1}$) |
|---|---|---|---|---|
| Literature | 10.3±0.5 | 7.2±0.4 | 140.4±0.5 | 7.5±1.0 |
| This work | 10.2±0.2 | 7.1±0.1 | 140.7±0.2 | 8.7±0.5 |

[a]References 2–6; Refs. 8 and 9; D. T. F. Marple, J. Appl. Phys. 35, 539 (1964); O. G. Lorimor and W. G. Spitzer, J. Appl. Phys. 36, 1841 (1965); D. Berlincourt, J. Jaffe, and L. R. Shiozawa, Phys. Rev. 129, 1009 (1963).

that for any function $f(\tau_k)$,

$$\langle f(\tau_k)\rangle = f(\langle \tau_k\rangle) \equiv f(\tau), \tag{8}$$

where the right-hand expression defines the quantity $\tau$. We note that under this assumption, $\epsilon_{fc}(\omega)$ is determined completely when $\omega_p^2$ and the drift mobility are known. Corrections to the assumption will be discussed later in this paper when comparisons are made between the electrical and optical determinations of carrier concentration and mobility.

In Fig. 1 we present the experimental reflectivity results and the computed best fits to the data assuming the validity of Eq. (8). The error bars represent one standard deviation in the data. The fits were made by means of a nonlinear least-squares-fit computer program.[14] In using the program, the only parameter assumed known was $m^*$. The program varied the four lattice parameters $\epsilon_s$, $\epsilon_\infty$, $\Gamma$, and $\omega_t$ and the free-carrier concentration and mobility in order to minimize $\chi^2$. The figure shows that the fits are uniformly good. Table II compares the values of the lattice parameters obtained from our fits with the means of the values found in the literature. The excellent agreement between the two sets of values and the fact that our fit values hardly varied from sample to sample is evidence for the validity of the theory used to fit the data and the fit procedure.

In Table I we compare the electrical and the optical values for the carrier concentrations and mobilities. In view of the facts that the Hall values have not been adjusted for deviations from unity of the Hall scattering factor and that we have used a simplified version of the optical theory, the agreement is good except for the lowest-concentration sample. To make a more valid comparison, we should include the low-field Hall scattering factor $r_H$, defined by[15]

$$r_H = \langle \tau_k^2\rangle / \langle \tau_k\rangle^2, \tag{9}$$

and also must take into account the averages given in Eq. (5).

Following the theory of Kukharskii and Subashiev[16] (hereafter referred to as KS), we treat the averaging by assuming that

$$\tau_k(\epsilon) \propto (\epsilon/kT)^s, \tag{10}$$

where $\epsilon$ is the carrier energy and $s$ is dependent on the scattering mechanism. It is true that the validity of Eq. (10) is in doubt in the case of polar mode optical scattering which is expected to be important in CdTe at 300 °K [10] but calculations by Ehrenreich,[17] in the Boltzmann limit, indicate that it is possible to define a temperature-dependent $s$ over a restricted range of temperatures. For CdTe at 300 °K, Ehrenreich's theory gives $s \approx 0.2$. At room temperature the only other important mode of scattering is by ionized impurities for which $s = \frac{3}{2}$.

KS introduce the functions $\gamma_1$ and $\gamma_2$ by defining

$$\left\langle \frac{\tau_k^2}{1+\omega^2\tau_k^2} \right\rangle = \gamma_1(\omega\tau, \eta, s)\, \frac{\tau^2}{1+\omega^2\tau^2} \tag{11}$$

and

$$\left\langle \frac{\tau_k}{1+\omega^2\tau_k^2} \right\rangle = \gamma_2(\omega\tau, \eta, s)\, \frac{\tau}{1+\omega^2\tau^2}, \tag{12}$$

where $\tau$ is defined in Eq. (8) and $\eta = \epsilon_F/kT$, where

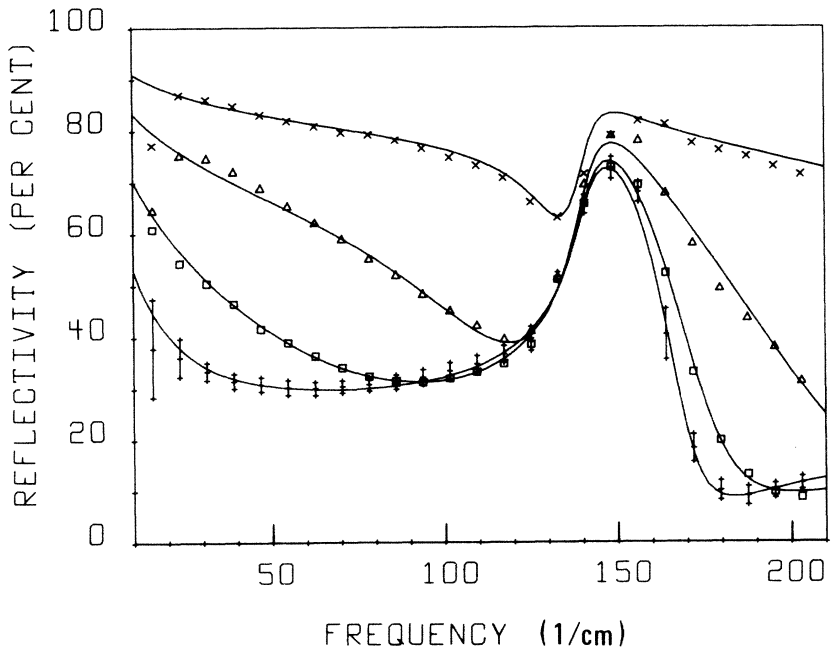


FIG. 1. Measured far-infrared reflectivity of $n$-CdTe vs frequency. The curves represent the least-squares fits to the data. + sample 9-71/2; □, 5-71/E; △, sample 32-70/I; ×, sample 4-70/A7. For clarity error bars are shown only for sample 9-71/2. Error bars for the other three samples are comparable.

$\epsilon_F$ is the Fermi energy. It can be shown that,[18] in the low-field limit,

$$r_H(H \to 0) = \gamma_1(0, \eta, s). \tag{13}$$

Thus, the theory of KS can be used to calculate scattering adjustments for both optical and Hall data. The calculation of the Hall scattering factor is not straightforward, since $r_H$ depends on $N$ through $\eta$ and an iterative or graphical procedure would be necessary to determine $N$ and $r_H$ in a self-consistent way.

In the case of the optical data the amount of computer time involved in fitting the data, if the proper averaging over the carrier distribution function is performed, is prohibitive. However, given a set of lattice and free-carrier parameters and a scattering mechanism, a calculation of the reflectivity spectrum according to the theory of KS is readily accomplished. If this reflectivity spectrum is then fitted with the conventional theory, a new set of carrier parameters is generated which can be compared with those which were used to calculate the KS reflectivity.

This has been done for ionized impurity scattering and for polar optical scattering in the following way. The values obtained from the Hall and resistivity measurements $N_H$ and $\mu_H$ were adjusted to obtain $N$ and $\mu$ by means of the scattering factor $r_H$ as determined from Eq. (13). These adjusted values were used in a calculation of the reflectivity spectrum according to the theory of KS. The spectrum generated from this theory was then fitted with the standard optical theory in order to determine the "optical" free-carrier parameters $N_{opt}$ and $\mu_{opt}$. In the case of polar optical mode scattering, the correction to $N_H$ and $\mu_H$ was in all cases less than 2% and to $N_{opt}$ and $\mu_{opt}$ less than $\frac{1}{2}$%. The corrections in the case of ionized impurity scattering were much more substantial and are detailed in Table III. To be realistic, one should consider mixtures of the two scattering mechanisms but it is evident from Table III that the agreement between the Hall and optical measurements of $N$ and $\mu$ would not be significantly improved by this refinement of the theory.

TABLE III. Effect of the inclusion of ionized impurity scattering on the electrical and optical determinations of carrier concentration and mobility. The values here have been adjusted for the scattering as described in the text and should be compared to the values in Table I.

| Sample | $r_H$ | Carrier concentration ($10^{18}$ cm$^{-3}$) Electrical | Optical | Mobility (cm$^2$/V sec) Electrical | Optical |
|---|---|---|---|---|---|
| 9-71/2 | 1.9 | 0.097 | 0.21 | 220 | 130 |
| 5-71/E | 1.9 | 0.27 | 0.21 | 360 | 380 |
| 32-70/I | 1.8 | 0.74 | 0.51 | 510 | 540 |
| 4-70/A7 | 1.6 | 2.1 | 1.5 | 470 | 570 |

One conspicuous feature of the reflectivity data in Fig. 1 is the appearance of well-defined minima whose position in frequency is a function of carrier concentration. These minima occur at the coupled plasmon-phonon frequencies, which, in the limit of infinite electron-scattering time and zero lattice damping are given approximately by[19]

$$2\omega_\pm^2 = (\omega_p^2 + \omega_l^2) \pm [(\omega_p^2 - \omega_l^2)^2 + 4\omega_p^2\omega_l^2(1 - \epsilon_\infty/\epsilon_s)]^{1/2}. \tag{14}$$

Here $\omega_l$, the longitudinal-optical lattice frequency, is given by

$$\omega_l^2 = (\epsilon_s/\epsilon_\infty)\,\omega_t^2. \tag{15}$$

For the four samples reported here, the Hall and resistivity measurements place $\omega_p$ in the neighborhood of $\omega_l$, hence the positions of the reflectivity minima should be strongly influenced by the plasmon-phonon interaction.

In the realistic case of finite $\tau_k$ and $\Gamma$ the solution for the coupled mode frequencies can be carried out by computer. We have carried out such a calculation for CdTe, using the simplifying assumption Eq. (8), and the results are given in Fig. 2 where curves are displayed for different drift mobilities. The effect of the finite scattering time is seen to be substantial.

For the lower-frequency branch of the plasmon-phonon interaction the dependence of the position of the minimum $\omega_-$ is primarily on the resistivity $\rho = m^*/Ne^2\langle\tau\rangle$ rather than on the carrier concentration and mobility separately. This results from two influences. At low carrier concentrations we have $\omega_p\tau_k \ll 1$ in which case the free-carrier part of the dielectric constant becomes

$$\epsilon_{fc}(\omega) \approx i\,4\pi/\omega\rho \tag{16}$$

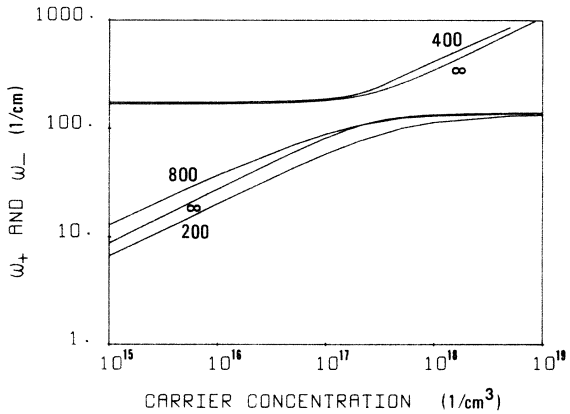


FIG. 2. Plasmon-phonon frequencies $\omega_+$ and $\omega_-$ vs carrier concentration in $n$-CdTe. The curves labeled $\infty$ are calculated for the case $\tau \to \infty$. The other curves are calculated for finite mobility values as indicated in units of cm$^2$/V sec.

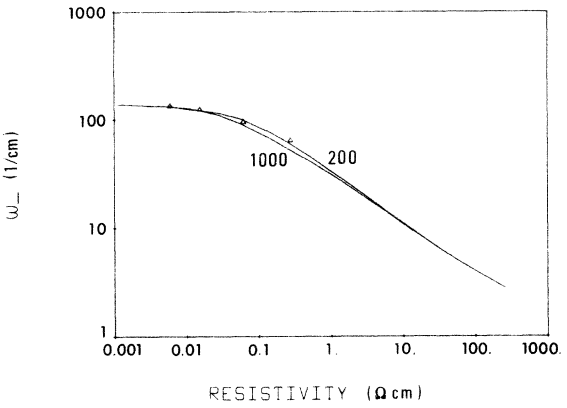


FIG. 3. Lower plasmon-phonon frequency $\omega_-$ vs resistivity in $n$-CdTe. The curves are labeled by the mobility values in $cm^2/V\,sec$. Triangles are experimental points.

near the low-frequency minimum. This result is valid regardless of the scattering model used. At higher carrier concentrations where $\omega_p\tau \gtrsim 1$ and for mobilities typical of CdTe, $\omega_p \gtrsim \omega_t$. In this case the position of the lower-frequency minimum tends to $\omega_t$ independently of the carrier concentration and the scattering behavior.

The dependence of $\omega_-$ on $\rho$ is shown in Fig. 3. The dependence on mobility is not completely absent, since the curves for $\mu = 200$ and $\mu = 1000$ $cm^2/V\,sec$ differ slightly. The variation with mobility is however much weaker than that displayed in Fig. 2. The experimental values of $\omega_-$ and $\rho$ are in good agreement with the theoretical curve.

Equation (16) suggests that for low-concentration samples it may be fruitless to try to find both carrier concentration and mobility from an analysis of far-infrared data, since $\epsilon_{fc}$ is almost completely determined by $\rho \propto 1/(N\mu)$. In Table I we compare the measured values of resistivity with optical values, defined by $\rho_{opt} = 1/(e N_{opt} \mu_{opt})$. The agreement is good and is striking in the case of the lowest concentration sample, where $N_{opt}$ and $\mu_{opt}$ are individually subject to large errors.

## CONCLUSIONS

The carrier concentrations and mobilities as determined from analysis of the reflectivity data are in fair agreement with those determined by Hall and resistivity measurements, at least for the three samples with the highest concentrations. Agreement between the Hall and optical measurements is not generally improved by inclusion of the more sophisticated scattering theory of KS. The optical data can be fitted quite well with the simple classical theory neglecting any energy dependence of the scattering time.

We can point to no specific reason why the use of the KS theory does not improve the agreement between the electrical values and the optical values of the free-carrier parameters. One possibility is that sample-to-sample variation in the degree of compensation may make our comparisons meaningless. A possible source of error lies in the fact that Ehrenreich's theory of a temperature-dependent $s$ for polar optical-mode scattering applies only to the Boltzmann limit while our samples are in a transition region. However, the indications are that the corrections due to this scattering mode are small and one would expect that in the degenerate limit the energy dependence of $\tau_k$ should become less important.

Despite the lack of data on the position of the high-frequency minimum for the two samples with the highest carrier concentrations, the influence of the plasmon-phonon interaction on the positions of the reflectivity minima has been clearly demonstrated for CdTe. The effect of finite carrier mobility on the position of the minima has been shown to be substantial. The lower frequency branch of the plasmon-phonon coupled mode is dependent primarily on resistivity rather than separately on $N$ and $\mu$. This behavior results essentially from the relatively low mobility of CdTe and is independent of the electron scattering mechanism.

The samples examined here at 300 °K are now being studied at lower temperatures. The mobility at lower temperatures should be influenced to a higher degree by ionized impurity scattering and effects due to the energy-dependent scattering time should be more pronounced. In addition the energy-dependent relaxation-time approximation for polar optical-mode scattering is expected to fail over a range of temperatures near 80 °K.

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