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UNIVERSITY OF ALBERTA

SOME ASPECTS OF ELECTRON BEHAVIOUR IN SOLIDS

BY

© W. J. HEANEY

A THESIS

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UNIVERSITY OF ALBERTA

FACULTY OF GRADUATE STUDIES AND RESEARCH

The undersigned certify that they have read, and
recommend to the Faculty of Graduate Studies and Research
for acceptance, a thesis entitled SOME ASPECTS OF ELECTRON
BEHAVIOUR IN SOLIDS, submitted by W.J. HEANEY in partial
fulfillment of the requirements for the degree of Doctor of
Philosophy.

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ABSTRACT

PART I

Two aspects of the behaviour of the electron gas
in the neighbourhood of an impurity in a metal are con-
sidered. In Chapter I it is shown that the fact that
a negative impurity is screened less effectively than
a positive impurity may be understood in terms of a
simple Thomas Fermi model. In Chapter II the collective
modes of oscillation of an electron gas in the neighbour-
hood of an impurity are investigated. A hydrodynamic
model is used in which the electrons are treated as a
charged Fermi fluid. It is shown that localized modes
of excitation exist near a positive impurity. Such a
local excitation can be detected in characteristic losses
of fast electrons.

PART II

Using the approach of Yamashita and Nakamura[25]
we consider the effect of induced phonons on the current
arising from an applied A.C. field in a piezoelectric
semiconductor. We restrict the calculation to the region
where the amplitude of the drift velocity of the electrons
is less than the velocity of sound. We find that for CdS
at $30^{\circ}K$ there is a critical frequency of the applied field
$\omega_c \sim 10^8$ sec^{-1}, such that the amplitude of the drift

current is constant for $\omega \ll \omega_c$ and drops to another constant value for $\omega \gg \omega_c$. We find that the current lags behind the applied field with a phase which has a maximum in the neighbourhood of ω_c.

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ACKNOWLEDGEMENTS

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PART I

INTRODUCTION

In part I of this thesis we consider some aspects of the behaviour of electrons in the presence of a fixed point impurity in a metal. In particular in Chapter I we treat the screening of an impurity by electrons, and in Chapter II, the collective modes of oscillation of the electrons near an impurity. We use a semi-classical model for the electrons, that is, we take account of the fact that they obey quantum statistics, but neglect all other quantum effects. We also neglect short range electron correlations and treat the lattice of the metal as if it formed a smeared out distribution of fixed positive charge. Thus we are using a conceptually simple model to describe the very complicated system of electrons in a metal. Indeed so many features of the real system are neglected that it might be expected that the model is totally inadequate. Yet this is not the case and for many purposes the model can be expected to give a good qualitative, even semi-quantitative description. To see why this is so we review the concepts that have been developed for treating interacting electrons in a metal.

The study of the free electron gas (i.e. electrons in a neutralizing fixed background of positive charge) began with Sommerfeld[1] in 1928. He treated the

electrons at zero degrees and neglected the Coulomb

interactions. Since interactions were neglected each

electron state could be labelled with a wave vector

k, which is related to the energy of the state

$\varepsilon_k = \hbar^2 k^2/2m$, where m is the electron mass. Sommerfeld

applied Fermi Dirac statistics to the electrons which

led to the introduction of the important concept of the

Fermi surface. This is a spherical surface of constant

energy in wave vector space which separates occupied

electron states from unoccupied states when the electron

gas is in its ground state. An excited state of the

system is created by transferring an electron from some

state k_1, below the Fermi surface to an unoccupied state

k_2, above the Fermi surface. The energy of the excited

state is then $\hbar^2 k_2^2/2m - \hbar^2 k_1^2/2m$. Such an energy spec-

trum is called a spectrum of the Fermi type.

Since that time many powerful techniques have been

developed to take into account the effects of particle

interactions.[2] One of the first theories to describe

interactions between fermions is due to Landau.[3] He

considered a system of fermions interacting via short

range forces (i.e. less than the order of an inter-

particle spacing) as a model for liquid He^3. Because

of interactions the concept of a one particle state no

longer has a well defined meaning. In an intuitive manner he

introduced the concept of a quasiparticle. This is a
fermion moving in the self-consistent short range field
of all the other fermions of the system. Thus the
energy of the quasi-particle depends on the distribu-
tion of other particles in the system. However he
postulated that each of the quasi-particle states could
be labelled with a wave vector $\underset{\sim}{k}$ and that they obey
Fermi Dirac statistics. The energy of a quasi-particle
state, however, is not related to the wave vector in a
simple fashion, since its energy depends on the distri-
bution of all the other particles. However, since the
quasi-particles obey Fermi Dirac statistics, there is
a Fermi surface and excitations completely analagous to
those of the Sommerfeld model. The quasi-particles of
the Landau theory are weakly interacting, with the
result that they have an appreciable lifetime only in
the neighbourhood of the Fermi surface and there is a
Fermi type spectrum only for weakly excited states.

It is often possible within solid state physics
to take account of interactions by defining an appro-
priate effective mass. This is only possible to a
limited extent within the Landau theory. Those expres-
sions which depend only on the density of states at
the Fermi surface, like the specific heat, can be taken
over from the Sommerfeld theory with the electron mass

replaced by an effective mass. However for example the expression for the pressure within the Landau theory differs from the non-interacting electron form by a term proportional to the density. In Chapter II we use the non-interacting electron gas value for the pressure. A more exact treatment would take into account the correction due to electron-electron inter- action.

So far we have only discussed the case of a Fermi gas interacting via short range forces. Silin[4] extended the theory of Landau to the electron gas, which is characterized by long range Coulomb forces. He found that it is possible to split the Coulomb force into a long range part and a short range part. He proceeded by considering a macroscopic disturbance in the density of the electron system. Since the electrons are charged the macroscopic disturbance of the electron density from equilibrium gives rise to a macroscopic electric field through Poissons equation. Silin showed that that part of the Coulomb interaction whose effects are not included in the macroscopic electric field are contained in short range screened Coulomb interactions between the electrons. Since the disturbance from equilibrium is on a macroscopic scale the system is assumed to remain uniform over a sufficiently large macroscopic region so that it is possible to define a

local distribution of electrons. Within the local distribution, the electrons are interacting via short range forces and it is possible to apply the Landau theory to them. The work of Landau and Silin was given a firm theoretical foundation by Galitski and Migdal.[5]

The macroscopic electric field introduced by Silin has another important effect besides being responsible for screening the interaction between electrons. It is also responsible for organizing the motion of the macroscopic disturbance of charge into well defied oscillations. That this effect should occur had been predicted four years earlier by Bohm and Pines[6] in 1952. They approached the problem of the Fermi gas interacting via Coulomb forces, by analogy with the classical plasma. A classical plasma is a high temperature overall neutral system containing charged particles, usually electrons and ions. It had been known for a long time that well defined density oscillations could be sustained in such a plasma,[20] and that the Coulomb field of a particle is screened by other particles.[21] Bohm and Pines looked for, and found similar behaviour in the interacting electron Fermi gas. They introduced a method for quantizing the density fluctuations and called the resulting quanta plasmons. These have a spectrum of the Bose type and have a dispersion

$$\omega_k^2 = \omega_o^2 + \frac{3}{5} v_F^2 k^2 + \frac{\hbar^2}{4m^2} k^4 + \ldots \qquad (1)$$

where $\omega_o^2 = 4\pi n e^2/m$; n is the electron number density and e is their charge. $\underset{\sim}{k}$ is the wave vector labelling one component of the Fourier decomposition of the oscillating electron density, v_F is the Fermi velocity.

The equation (1) holds only for small values of $\underset{\sim}{k}$. This is easy to understand since the plasmon corresponds to fluctuations in the electron density and a fluctuation must contain at least one particle. Roughly we must have $k \lesssim \frac{1}{a}$ where a is the interparticle spacing. Since $\frac{3}{5} v_F^2$ is the average square of the velocity, we note that in equation (1) terms containing h do not occur until order k^4. This suggests that the first two terms in equation (1) may be obtained using a model which neglects quantum effects, except for the statistics. Powell[19] has found that the peak in the energy loss spectrum of k.e.v electrons is predicted quite well by the free electron value of ω_o.

The picture that emerges of the free electron gas from the work of the above authors and many others[2] has much in common with the picture of the non-interacting gas of Sommerfeld. There exists a spherical Fermi surface and there are weak excitations of the single particle Fermi type, though these particles are no longer bare electrons. In addition the interacting

electron gas has a collective behaviour, not shared
with the non-interacting system, which is connected
with the long range part of the Coulomb interaction.
These collective excitations have a spectrum of the
Boase type. The remaining short range part of the
Coulomb interaction gives rise to short range electron
correlations. These two types of behaviour are not
independent and are weakly coupled together in a com-
plicated manner.

Much of the impetus for the work on the free
electron gas came from the desire to understand the
behaviour of electrons in a real metal. The investi-
gation of the behaviour of electrons in a real metal
has to take into account the effects of the interaction
of the electrons with the crystal lattice. Electron
states in a periodic lattice can be labelled with a
wave vector $\underset{\sim}{k}$, in much the same way that free electron
states can be. However this wave vector is not related
to the momentum, and the energy of a state may be a
complicated function of the wave number even if the
Coulomb interaction between electrons is neglected.
The periodic lattice gives rise to bands of states,
which the electrons can occupy, separated by energy
gaps where there are no states available for occupancy.
In a metal at zero temperature the highest band in

energy, the conduction band is partially occupied by
electrons, and all other bands of lower energy are
full. Thus in wave vector space all available electron
states are occupied up to a surface of constant energy.
This surface is the Fermi surface for the electrons in
a metal. It need not be spherical in shape, as is the
case for the free electron gas.

In 1957 Pippard[7] measured and plotted the Fermi
surface of copper. Soon studies of the geometry of the
Fermi surface of other metals were done and showed that
for a large number of metals the Fermi surfaces were
remarkably close to the spherical surface of a free
electron gas. Thus it was realized that the periodic
potential of the lattice had a relatively small effect
on the electrons in a metal, outside of creating energy
gaps. The free electron nature of the Fermi surfaces
suggests that the electron states in metals might be
obtainable through perturbation theory from the free
electron states. These considerations led to the deve-
lopment of the pseudo-potential method[8] for applica-
tion to electrons in metals. However because of the
complexity of the calculations, and the "non-physical"
nature of the pseudo-potentials, the free electron
model still remains a valuable source of understanding
of the behaviour of the electrons in a metal. It may

be expected to give a good qualitative description of electronic properties of those metals in which the Fermi surface is almost spherical.

We consider two problems concerning the behaviour of the electron gas in the neighbourhood of a fixed point impurity. In Chapter I we consider the screening of the impurity by the electrons and in Chapter II we look for collective modes of oscillation of the electrons localized near the impurity. As we have mentioned screening and collective behaviour are brought about by the long range part of the Coulomb interaction. In Chapters I and II we consider only the long range part of the Coulomb interaction and neglect the short range correlations between electrons. In fact we do not consider the particle nature of the electrons at all. For if one neglects the short range correlations, but takes into account the random motion of the electrons, one finds that this exhibits itself in damping of the collective modes.[18] At metallic densities effects of short range correlations, and random motion are expected to be of importance.[17] We have neglected both these effects and cannot expect to achieve quantitative agreement. However our results should be applicable in a qualitative manner to several metals.

First we discuss a time independent steady state problem of the screening of an impurity by the electron

gas. It is well known from pseudo-potential calcula-
tions[9] that a positive impurity in a metal is
screened more effectively than a negative impurity
we show that this effect may be qualitatively under-
stood by an elementary argument based on the Thomas
Fermi method.

We next treat the problem of the collective
excitations of the electron gas in the presence of
an impurity. Single particle excitations of the
electron gas in the presence of an impurity have been
investigated by Layzer.[10] He has shown that in the
vicinity of a positive impurity localized single par-
ticle excitations exist. The collective modes of
oscillation of an electron gas in the presence of an
impurity have been studied by Sziklas[11] and Sham[12].
These authors have used a quantum mechanical many-body
approach, and have found that plasmon type excitations
localized in the vicinity of a negative impurity exists
with frequency $\sim \omega_0/\sqrt{2}$. ω_0 is the plasma frequency of
a homogeneous medium. Sziklas[11] has concluded that
no localized excitation exists in the neighbourhood of
a positive impurity. These authors have also used
crude hydrodynamic models to provide a qualitative
check of the results of their microscopic theories.
One of the problems with the many body approach is that
while the mathematical nature of the assumptions is

clear enough, the physical nature of these assumptions
is not. For instance Sham in his investigation, ex-
pands the dielectric function for the non-uniform
electron gas in powers of a certain parameter, and in
a mathematically consistent manner neglects higher
order terms in the expansion. However hidden among
these higher order terms, are terms of physical impor-
tance. In a systematic hydrodynamic model, these extra
terms occur in a straightforward manner.

In Chapter II, we present a hydrodynamic model
of the electron gas in the neighbourhood of a positive
impurity in a metal. We find that localized collective
modes exist with frequency $\omega \sim \sqrt{3}\, \omega_o$.

CHAPTER I

SCREENING OF A NEGATIVE IMPURITY IN METALS

We consider the screening of a fixed point impurity by the free electron gas. We denote by $V(r)$ the potential at point r due to the impurity and the non-uniform induced charge density. We consider a volume at the point r which is macroscopically small, but microscopically large so that it contains a large number of particles. If we neglect the Coulomb interaction between these electrons the electron states in the small volume at point r can be labelled with a wave vector k. This procedure is legitimate only if the potential $V(r)$ varies slowly over distances of the order of an interparticle spacing. Close to the impurity this is not the case. However we use this picture only to obtain the potential at distances far from the impurity. Close to the impurity the potential is obtained by different methods. The two potentials are then matched in a plausible fashion.

In the ground state at $T = 0$ the electron states in the small volume at r, are occupied to a maximum value of k, $k_F(r)$ which is a function of r. We could take into account the effect of interactions by using the quasi-particle picture here. They have the same Fermi surface as the non-interacting electron gas.

However as mentioned in the introduction the most important factor determining screening is the long range part of the Coulomb interaction which is given by $V(\underset{\sim}{r})$. The electrons at the point $\underset{\sim}{r}$ in addition to their kinetic energy $h^2k^2/2m$ have a potential energy $-eV(\underset{\sim}{r})$. In the ground state of the total system in equilibrium we then have

$$\frac{h^2k_F^2(r)}{2m} - eV(r) = \mu \qquad (1.1)$$

where μ is the chemical potential of the total system. Under the assumption that $V(\underset{\sim}{r}) \to 0$ as $r \to \infty$ we have

$$\mu = \epsilon_F = \frac{h^2(3\pi^2)^{2/3}}{2m} \, n_o^{2/3} \qquad (1.2)$$

where n_o is the electron number density at very large distances from the impurity. Equation (1.1) is the condition for equilibrium in the ground state. For if $h^2k_F^2(r)/2m > \mu + eV(r)$ then an electron with such a kinetic energy could escape to infinity where it would be above the Fermi surface there.

The number of electrons per unit volume in the small volume at $\underset{\sim}{r}$ is

$$n(\underset{\sim}{r}) = \frac{2}{(2\pi)^3} \int_0^{k_F(\underset{\sim}{r})} d^3k = \frac{4}{(2\pi)^2} \frac{k_F^3(\underset{\sim}{r})}{3} \quad . \qquad (1.3)$$

Substituting (1.3) into (1.1) we obtain the well known Thomas-Fermi expression for the number density in terms of the potential

$$n(\underset{\sim}{r}) = \frac{1}{3\pi^2} \frac{2m}{\hbar^2} \{(\varepsilon_F + eV(\underset{\sim}{r}))\}^{3/2} \quad . \tag{1.4}$$

Poisson's equation is

$$\nabla^2 V(\underset{\sim}{r}) = 4\pi e n(\underset{\sim}{r}) \quad .$$

Using Poisson's equation together with (1.4) we can solve for $V(\underset{\sim}{r})$. We assume that $V(r)$ is spherically symmetric. Then at large distances from the impurity such that $\left|\frac{eV(r)}{\varepsilon_F}\right| < 1$ the physically acceptable solution is given by[13]

$$V(r) = \frac{C_1 e^{-qr}}{r} \tag{1.6}$$

where C_1 is a constant and

$$q^2 = \frac{4me^2}{\hbar^2} \left(\frac{3n_o}{\pi}\right)^{1/3} \quad . \tag{1.7}$$

The constant C_1 is to be determined by requiring that equation (1.6) matches with the potential at small distances from the impurity. Close to the impurity where $\left|\frac{eV(r)}{\varepsilon_F}\right| > 1$ we treat the positive and negative impurities separately.

Positive Impurity

At very close distances to the impurity the potential goes as Ze/r where Z is positive. The solution (1.6) has the required form of a Coulomb potential for qr << 1. Therefore

$$V(r) = \frac{Ze}{r} e^{-qr} \tag{1.8}$$

is a good approximation to the actual form of the potential for all values of r.

Negative Impurity

A positive impurity attracts electrons to itself which screen its charge. On the other hand a negative impurity repels electrons. Since the potential $V(r)$ in this case is negative at some point $r = r_o$,

$$\varepsilon_F + eV(r_o) = 0 . \tag{1.9}$$

In this case where $r < r_o$ the Thomas-Fermi equation (1.4) is not even approximately valid since it gives imaginary values for the electron charge density. Hence we assume that for $r < r_o$, $n(r) = 0$. Then by solving Poisson's equation within this region, $r < r_o$, we obtain

$$V(r) = -\frac{2}{3} \pi e n_o r^2 - \frac{Ze}{r} + C_2 \tag{1.10}$$

where C_2 is a constant and Z is positive. For $r > r_o$ equation (1.6) may still be regarded as approximately valid except that the constant C_1 is as yet unknown. We determine C_1 and C_2 by requiring that the potential and its gradient be continuous at $r = r_o$. One then obtains

$$V(r > r_o) = - e \left\{ \frac{(Z - \eta^3)}{\eta q a + 1} \frac{e^{-q(r-\eta a)}}{r} \right\} \qquad (1.11)$$

and

$$V(r < r_o) = - \frac{Ze}{r} - \frac{1}{2} \frac{er^2}{a^3} + \frac{e}{\eta a} \left[\frac{1}{2} \eta^3 (1 + \frac{2}{1+q\eta a}) + \frac{Zqa\eta}{1+qa\eta} \right]$$

$$(1.12)$$

where $\eta = r_o/a$, $\frac{4}{3} \pi a^3 n_o = 1$.

Within the framework of our model η may be found by substituting (1.11) or (1.12) into (1.9). We obtain

$$\eta^3 + \alpha q a \eta^2 + \alpha \eta - Z = 0 \qquad (1.13)$$

where $\alpha = \varepsilon_F/(e^2/a)$. At an electron density of 10^{23} electrons/c.c. $a \sim 2a_o$ where a_o is the Bohr radius; hence $e^2/a \sim 13$ eV. In many metals $\varepsilon_F \sim 5$ eV, thus $\alpha \sim 0.5$. In metals $q \sim \frac{1}{a_o}$ and therefore $qa \sim 2$. Hence for $Z = 1$ equation (1.13) has a solution, $\eta \sim 0.7$. Thus a negative impurity with $Z = 1$ creates a hole of radius of the order of an inter-electron spacing around itself in a metal. The radius of this hole is

larger than the range of the field of a positive
impurity. The electrons in a metal therefore see
a larger cross section due to a negative impurity
than that due to a positive impurity. This difference
should exhibit itself in the residual resistance.

We may also consider the impurity as if it is
an electron itself. Equations (1.11) and (1.12)
then give the field of a slowly moving electron in
an electron gas. In the limit of high electron density
(i.e. much higher than metallic densities) equation
(1.13) has the solution $\eta \sim 0$. In this limit equations
(1.11) and (1.12) reduce to the well known form of the
screened Coulomb interaction

$$V(r) \approx - e \frac{e^{-qr}}{r} \quad , \qquad n \to 0 \quad . \tag{1.14}$$

However at metallic densities (1.11) and (1.12) should
be a better form of the short range Coulomb interaction
between electrons.

As an example we calculate the residual resis-
tances in copper due to negative and positive impu-
rities. The Fermi surface in copper is not even
topologically equivalent to a sphere since the surface
intersects the zone boundaries. However the area in
contact with the zone boundary is small compared to
the area of the total Fermi surface. Neglecting the

effects of the contact of the Fermi surface and the
zone boundary, a spherical Fermi surface should be a
qualitatively good approximation to the actual Fermi
surface in copper. For small impurity concentrations
the residual resistance per impurity atom, ρ, is given
by the well known expression[13]

$$\rho = B \int_0^\pi (1 - \cos\theta)\sin\theta \; I(\theta)d\theta \qquad (1.15)$$

where $I(\theta)$ is the differential scattering cross section
of the impurity. Since the impurity potential is
spherically symmetric I does not depend on the
asmuthal angle. B is a constant. In the Born appro-
ximation, the differential cross section for scattering
from an incident state $\underset{\sim}{k}$ to a final state $\underset{\sim}{k}'$ is propor-
tional to

$$M_{\underset{\sim}{k},\underset{\sim}{k}'} = \left| \int e^{-i\underset{\sim}{k}\cdot\underset{\sim}{r}} \; V(r) \; e^{i\underset{\sim}{k}'\cdot\underset{\sim}{r}} \; d^3r \right|^2 \quad . \qquad (1.16)$$

For the positive impurity the matrix element for
scattering in the Born approximation is well known[13]
and given by

$$M_{\underset{\sim}{k},\underset{\sim}{k}'}^+ = e \int e^{-i\underset{\sim}{k}\cdot\underset{\sim}{r}} \; V_+(r) e^{i\underset{\sim}{k}'\cdot\underset{\sim}{r}} \; d^3r$$

$$= - \frac{4\pi Z e^2}{|\underset{\sim}{k}-\underset{\sim}{k}'|^2 + q^2} \qquad (1.17)$$

where $V_+(r)$ is given by (1.8). In the case of a nega-
tive impurity the matrix element for scattering in the
Born approximation is

$$M^-_{\underset{\sim}{k},\underset{\sim}{k}'} = -e \int_0^{r_o} e^{-i\underset{\sim}{k}\cdot\underset{\sim}{r}} \, V_-(r<r_o) \, e^{i\underset{\sim}{k}'\cdot\underset{\sim}{r}} \, d^3r$$

$$-e \int_{r_o}^\infty e^{-i\underset{\sim}{k}\cdot\underset{\sim}{r}} \, V_-(r>r_o) \, e^{i\underset{\sim}{k}'\cdot\underset{\sim}{r}} \, d^3r \quad . \qquad (1.18)$$

Using (1.11) and (1.12) we obtain:

$$M^-_{\underset{\sim}{k},\underset{\sim}{k}'}(r<r_o) = \frac{4\pi Ze^2}{|\underset{\sim}{k}-\underset{\sim}{k}'|^2} \left[1 + \left(\frac{3\eta}{a^2|\underset{\sim}{k}-\underset{\sim}{k}'|^2} + \frac{\eta^3}{1+\eta qa} + \frac{\eta qa}{1+\eta qa} \right. \right.$$

$$\left. - 1 \right) \cos\eta a|\underset{\sim}{k}-\underset{\sim}{k}'| + \left(\frac{\eta^2}{a|\underset{\sim}{k}-\underset{\sim}{k}'|}[1 - \frac{1}{\eta qa}] - \frac{3}{a^3|\underset{\sim}{k}-\underset{\sim}{k}'|^3} \right.$$

$$\left. - \frac{1}{|\underset{\sim}{k}-\underset{\sim}{k}'|^2} \frac{q^2}{(1+\eta qa)} \right) \sin\eta a|\underset{\sim}{k}-\underset{\sim}{k}'| \left. \vphantom{\frac{1}{1}} \right] \qquad (1.19)$$

$$M^-_{\underset{\sim}{k},\underset{\sim}{k}'}(r>r_o) = \frac{4\pi Ze^2}{|\underset{\sim}{k}-\underset{\sim}{k}'|^2} \frac{\eta^3 - Z}{1+\eta qa} \{ |\underset{\sim}{k}-\underset{\sim}{k}'| \cos\eta a|\underset{\sim}{k}-\underset{\sim}{k}'|$$

$$+ q \sin\eta a|\underset{\sim}{k}-\underset{\sim}{k}'| \} \{ |\underset{\sim}{k}-\underset{\sim}{k}'|^2 + q^2 \}^{-1} \quad . \qquad (1.20)$$

Recalling that the differential cross section is
proportional to (1.16) we can substitute for $I(\theta)$ in

(1.15) from (1.17), (1.19) and (1.20), and obtain the residual resistances per impurity atom. The ratio of the residual resistances per impurity atom, for positive and negative impurities of the same concentration with $Z = 1$ is

$$\rho_+/\rho_- = \left[\int_0^1 dx\ x^3(x^2+b_2^2)^{-2}\right] \times \left(\left[\int_0^1 dx\ x^3 \left(\frac{1}{x^2}\ (1\right.\right.\right.$$

$$+ \ [\frac{3}{b_1^2 x^2} + \frac{\eta^3}{1+\eta qa} + \frac{\eta qa}{1+\eta qa} - 1]\ \cos b_1 \eta x$$

$$+ \ [\frac{\eta^2}{b_1 x}\ (1 - \frac{1}{\eta qa}) - \frac{3}{b_1^3 x^3} - \frac{b_2}{x(1+\eta qa)}]\ \sin b_1 \eta x$$

$$+ \ \frac{1}{x}\ (\frac{1-\eta^3 x}{1+\eta qa}\ [\frac{x\ \cos b_1 \eta x + b_2\ \sin b_1 \eta x}{x^2 + b_2^2}]))\right]^2 \Big)^{-1}$$

$$\tag{1.21}$$

where $b_1 = 2(\frac{9\pi}{4})^{1/3}$, $b_2 = q/2k_F$.

In deriving (1.21) we have assumed elastic scattering so that $|\underset{\sim}{k}-\underset{\sim}{k}'| = 2k_F \sin \theta/2$ where θ is the angle between $\underset{\sim}{k}$ and $\underset{\sim}{k}'$. We have made the substitution $x = \sin \theta/2$.

We obtain $\eta = 0.8$ from equation (1.13) for a single negative impurity in copper, where we have used

the mass of the free electron and assumed one free electron per copper atom. Substituting $\eta = 0.8$ in equation (1.21) we find $(\rho_+/\rho_-) = 0.25$. We may note that as expected from the above treatment ρ_+/ρ_- is less than unity for other sets of impurities in copper (see reference (9(b)) page 341). The resistivity measurements in Ag, Pd and Pt with impurities of neighbouring elements in the periodic table exhibits similar behaviour[14]. However the case of Ni and Zn as hosts are an exception.[15]

The results given here demonstrate how a very simple model can give reasonable qualitative agreement with experimental data on complicated systems, Ag, Cu, Pd and Pt are not simple metals in the sense that the alkalis are, however experimental data on the alkalis is not available, presumably because of the difficulty of inserting impurities.

Comment

After the material contained in Chapter I had been accepted for publication it came to my attention that the same problem had been treated in a very similar fashion by Alfred and March in 1956.[22]

CHAPTER II

COLLECTIVE MODES OF OSCILLATION OF AN ELECTRON
GAS IN THE VICINITY OF AN IMPURITY IN METALS

In this chapter we investigate the behaviour of the collective modes of vibration of the electron gas in the neighbourhood of an impurity at zero degrees. We regard the electrons as a charged Fermi fluid, and determine the motion of the density fluctuations due to electric and pressure forces through Euler's equation and the equation of continuity.

The electrons and a fixed point impurity of charge Ze are enclosed in a box of volume Ω. The total system is electrically neutral due to the presence of a smeared out fixed uniform background of positive charge. In equilibrium the electron number density $\rho(\underset{\sim}{r})$ will assume some stationary space dependent form, which we expand in a Fourier series:

$$\rho(\underset{\sim}{r}) = \sum_{\underset{\sim}{k}} \rho_{\underset{\sim}{k}} \, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \quad . \tag{2.1}$$

Assuming periodic boundary conditions the values of k in the summation in (2.1) are given by

$$\underset{\sim}{k} = \frac{2\pi}{L} \, (n_x\underset{\sim}{i} + n_y\underset{\sim}{j} + n_z\underset{\sim}{k})$$

where n_x, n_y, and n_z are integers and $L^3 = \Omega$. Hence it follows that

$$\frac{1}{\Omega} \int_{\Omega} e^{i(\underset{\sim}{k}-\underset{\sim}{q})\cdot\underset{\sim}{r}} \, d^3r = \delta_{\underset{\sim}{k},\underset{\sim}{q}} \qquad (2.2)$$

and from (2.1) and (2.2)

$$\rho_{\underset{\sim}{k}} = \frac{1}{\Omega} \int_{\Omega} \rho(\underset{\sim}{r}) \, e^{-i\underset{\sim}{k}\cdot\underset{\sim}{r}} \, d^3r \qquad (2.3)$$

Associated with the equilibrium charge density is an electric field $\underset{\sim}{E} = -\text{grad } \Phi$ where Φ is given by Poisson's equation

$$\nabla \cdot E = -\nabla^2 \Phi = -4\pi e\{\rho(\underset{\sim}{r}) - Z\delta(\underset{\sim}{r}) - \rho^+\} \qquad (2.4)$$

where e is the magnitude of the electron charge, Ze is the charge on the impurity and ρ^+ is the uniform positive charge number density. Expanding the potential as

$$\Phi(\underset{\sim}{r}) = \sum_{\underset{\sim}{k}} \phi_{\underset{\sim}{k}} \, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \qquad (2.5)$$

and substituting (2.1) and (2.5) into (2.4) we obtain

$$\sum_{\underset{\sim}{k}} k^2 \phi_{\underset{\sim}{k}} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} = -4\pi e\{ \sum_{\underset{\sim}{k}} \rho_{\underset{\sim}{k}} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} - Z \sum_{\underset{\sim}{k}} \frac{1}{\Omega} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} - \rho^+\}$$

$$(2.6)$$

where the Fourier component of the δ function is $1/\Omega$, which follows from (2.3). The fact that the total system is electrically neutral means

$$- e \int_{\Omega} \rho(\underset{\sim}{r}) d^3 r = -e\Omega\rho_o = -\{Ze + \rho^+ \Omega e\} \quad . \qquad (2.7)$$

Hence using (2.7) (2.6) can be written

$$\sum_{\underset{\sim}{k}} k^2 \phi_{\underset{\sim}{k}} = \sum_{\underset{\sim}{k}\neq 0} + \frac{4\pi e}{k^2} \{\frac{Z}{\Omega} - \rho_{\underset{\sim}{k}}\} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \quad . \qquad (2.8)$$

Thus

$$\phi_{\underset{\sim}{k}} = \frac{4\pi e}{k^2} \{\frac{Z}{\Omega} - \rho_{\underset{\sim}{k}}\} \qquad \underset{\sim}{k} \neq 0$$

$$\phi_{\underset{\sim}{k}} = 0 \qquad \underset{\sim}{k} = 0 \quad . \qquad (2.9)$$

If the electron density is displaced slightly from equilibrium and then left free to move under the influences of the forces in the system, a small fluctuating charge density $- e\delta\rho(\underset{\sim}{r},t)$ is thereby induced in the system. We expand the fluctuating charge number density in a Fourier series

$$\delta\rho(\underset{\sim}{r},t) = \sum_{\underset{\sim}{k}\neq 0} \sigma_{\underset{\sim}{k}}(t) e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \quad . \qquad (2.10)$$

We are interested in finding the normal modes of vibration of the density fluctuations. In a uniform electron gas, to a good approximation for small values of $\underset{\sim}{k}$, each Fourier component of the density fluctuation oscillates independently with a frequency $\omega(\underset{\sim}{k})$. Because the system we are considering is spatially non-uniform

in equilibrium, it is to be expected that each normal mode will have a spatial structure which will be especially pronounced for the localized modes. If the normal modes of the density fluctuation have a spatial structure many Fourier components are required to build this structure. Hence we look for density fluctuations of the system that have the form

$$\delta\rho(\underset{\sim}{r},t) = e^{i\omega t} \sum_{\underset{\sim}{k}\neq 0} \sigma_{\underset{\sim}{k}}(\omega) e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \quad . \tag{2.10c}$$

The time dependent charge fluctuation (2.10a) produces a potential $\psi(\underset{\sim}{r},t)$,

$$\psi(\underset{\sim}{r},t) = e^{i\omega t} \sum_{\underset{\sim}{k}\neq 0} \psi_{\underset{\sim}{k}}(\omega) \, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \tag{2.11}$$

where from Poisson's equation

$$\psi_{\underset{\sim}{k}} = - \frac{4\pi e}{k^2} \sigma_{\underset{\sim}{k}} \quad . \tag{2.12}$$

$\Phi(\underset{\sim}{r})$ and $\psi(\underset{\sim}{r},t)$ together determine the electric field in the system.

We now consider the pressure forces. We know that in the presence of the impurity the electrons assume a spatially non-uniform distribution. This non-uniform distribution gives rise to an electric field and hence to electric forces which act on the electrons. We can

use the fact that in equilibrium the electric force on a local element of charge density must be balanced by other forces to define a pressure gradient. The total electric force on the electrons in a small volume V at a point $\underset{\sim}{r}$ is

$$\int_V e\rho(\underset{\sim}{r})\text{grad}\Phi(\underset{\sim}{r})d^3r$$

where from equation (1.1)

$$e\Phi(\underset{\sim}{r}) = \varepsilon_F(\underset{\sim}{r}) - \mu \quad . \tag{2.13}$$

From equation (1.3)

$$\varepsilon_F(\underset{\sim}{r}) = F[\rho(\underset{\sim}{r})]^{2/3} \tag{2.14}$$

where

$$F = (\hbar^2/2m)(3\pi^2)^{2/3} \quad . \tag{2.15}$$

Using (2.13) and (2.14) the electric force is

$$F \int_V \rho(\underset{\sim}{r})\text{grad } \rho^{2/3}(\underset{\sim}{r})d^3r \quad .$$

The force on the electrons due to the pressure $p_o(\underset{\sim}{r})$ is

$$-\int_S p_o(\underset{\sim}{r}) \underset{\sim}{n}_o dS$$

where S is the surface bounding V and $\underset{\sim}{n}_0$ is the outward
unit normal to S. In equilibrium the electric and
pressure forces balance so that

$$F \int_V \rho(\underset{\sim}{r})\text{grad } \rho^{2/3}(\underset{\sim}{r})d^3r = \int_S p_0(\underset{\sim}{r})\underset{\sim}{n}_0 \text{ dS}$$

$$= \int_V \text{grad } p_0(\underset{\sim}{r})d^3r \quad (2.16)$$

Choosing V small enough so that $p(\underset{\sim}{r})$ and $\rho(\underset{\sim}{r})$ do not
vary much over V we obtain from (2.16)

$$\text{grad } p_0(\underset{\sim}{r}) = \frac{2}{5} F \text{ grad } \rho^{5/3}(\underset{\sim}{r}) \quad . \quad (2.17)$$

Thus

$$p_0(\underset{\sim}{r}) = \frac{2}{5} F \rho^{5/3}(\underset{\sim}{r}) \quad (2.18)$$

since the constant of integration has no physical sig-
nificance and can be chosen as zero.

Equation (2.18) gives the pressure as a function
of the density for the electrons in equilibrium. Under
non-equilibrium conditions the resultant of the elec-
tric and pressure forces is not zero and it produces
local accelerations and hence vibrations of the fluid.
At low frequencies of vibration one expects that equa-
tion (2.18) would be a good expression for the pressure
if the time dependent density was substituted for $\rho(\underset{\sim}{r})$.

However at high frequencies characteristic of plasma oscillations the expression for the pressure should be modified from that at equilibrium.[12,16] We assume that in the high frequency case the pressure $p(r,t)$ is given by

$$p(r,t) = \frac{18}{25} F\, n^{5/3}(r,t) \quad, \tag{2.19}$$

where $n(r,t)$ is the time dependent electron number density

$$n(r,t) = \rho(r,t) + \delta\rho(r,t) \quad. \tag{2.20}$$

The justification for (2.19) is that this form of the pressure leads to a dispersion relation for the plasma oscillations of a uniform system that agrees with the one obtained by a microscopic treatment.

The equation of motion for a small element of volume V of the electron fluid acted upon by the electric and pressure forces is

$$\frac{d}{dt} m \int_V n(r,t)\,(r,t)d^3r = - \int_S p(r,t)n_0 dS$$

$$+ e \int_V n(r,t)grad(\Phi(r) + \psi(r,t)d^3r \tag{2.21}$$

where m is the electron mass. The element of volume V
moves with velocity $\underset{\sim}{v}(\underset{\sim}{r},t)$ such that the total mass
included is constant. Thus $mn(\underset{\sim}{r},t)d^3r = dm$ is indepen-
dent of time. Equation (2.21) can therefore be written,
on letting the volume V approach zero, as

$$mn \frac{dv}{dt} = -\text{grad } p(t) + en \text{ grad}(\Phi + \psi) \quad . \qquad (2.22)$$

Equation (2.22) is the Euler form of the equation of
motion for an electron fluid.

The equation of continuity is

$$\frac{\partial n}{\partial t} + \text{div}(n\underset{\sim}{v}) = 0 \quad . \qquad (2.23)$$

In equilibrium $\underset{\sim}{v} = 0$, $\delta\rho = 0$ and $\psi = 0$. We treat these
as first order small quantities. Combining equations
(2.22) and (2.23) and keeping terms linear in V and $\dot{n}$
we obtain

$$m \frac{\partial^2 n}{\partial t^2} = \text{div}[\text{grad } p(t) - en \text{ grad}(\Phi + \psi)] \quad . \quad (2.24)$$

Substituting for $p(t)$ and n in equation (2.24)
from (2.19) and (2.20) and keeping terms linear in
$\delta\rho(\underset{\sim}{r},t)$ and $\psi(\underset{\sim}{r},t)$ we have

$$m \frac{\partial^2}{\partial t^2} \delta\rho(\underset{\sim}{r},t) = \text{div grad}\{F[\rho(\underset{\sim}{r})]^{5/3} \frac{18}{25}[1 + \frac{5}{3} \frac{\delta\rho(\underset{\sim}{r},t)}{\rho(\underset{\sim}{r})}]$$

$$- e\,\text{div}\{\rho(\underset{\sim}{r})\text{grad}(\Phi + \psi) + \delta\rho(\underset{\sim}{r},t)\text{grad } \Phi\} \quad . \qquad (2.25)$$

The time independent part of (2.25) is not to be considered as the condition for equilibrium in view of the meaning given to $p(\underset{\sim}{r},t)$. The time dependent part of (2.25) gives

$$m \frac{\partial^2}{\partial t^2} \delta\rho(\underset{\sim}{r},t) = \text{div grad}\{\frac{9}{5} F[\rho(\underset{\sim}{r})]^{2/3} \delta\rho(\underset{\sim}{r},t)\}$$

$$- e \text{ div } \{\rho(\underset{\sim}{r}) \text{ grad } \psi + \delta\rho(\underset{\sim}{r},t) \text{ grad } \phi\} . \qquad (2.26)$$

We expand $[\rho(\underset{\sim}{r})]^{2/3}$ as

$$[\rho(\underset{\sim}{r})]^{2/3} \approx \rho_o^{2/3} [1 + \sum_{\underset{\sim}{k}\neq 0} \frac{\rho_k}{\rho_o} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}] \qquad (2.27)$$

Substituting (2.27) into (2.26) and taking the Fourier transform of (2.26) we obtain

$$\frac{\omega^2}{\omega_o^2} \sigma_{\underset{\sim}{q}} = \frac{9}{5} \frac{q^2}{k_o^2} \sigma_{\underset{\sim}{q}} + \sigma_{\underset{\sim}{q}} + \sum_{\underset{\sim}{k}'\neq\underset{\sim}{q}} \frac{q-k'}{k'^2} \frac{\rho_{\underset{\sim}{q}-\underset{\sim}{k}'}}{\rho_o} \sigma_{\underset{\sim}{k}'}$$

$$+ \sum_{\underset{\sim}{k}'\neq\underset{\sim}{q}} \frac{\underset{\sim}{q}\cdot(\underset{\sim}{q}-\underset{\sim}{k}')}{|\underset{\sim}{q}-\underset{\sim}{k}'|^2} (\frac{\rho_{\underset{\sim}{q}-\underset{\sim}{k}'}}{\rho_o} - \frac{z}{\rho_o\Omega}) \sigma_{\underset{\sim}{k}'}$$

$$+ \frac{9}{5} \frac{2}{3} \sum_{\underset{\sim}{k}'\neq\underset{\sim}{q}} \frac{q^2}{k_o^2} (\frac{\rho_{\underset{\sim}{q}-\underset{\sim}{k}'}}{\rho_o}) \sigma_{\underset{\sim}{k}'} \qquad , \qquad (2.28)$$

where

$$\omega_o^2 = 4\pi\rho_o e^2/m$$

$$k_o^2 = \frac{4\pi\rho_o e^2}{\frac{2}{3} F\rho_o^{2/3}} \qquad .$$

In obtaining (2.28) we have substituted for $\rho(\underset{\sim}{r})$, $\Phi(\underset{\sim}{r})$, $\delta\rho(\underset{\sim}{r},t)$, $\psi(\underset{\sim}{r},t)$ from equations (2.1), (2.5), (2.10a) and (2.11), and made use of equations (2.9) and (2.12) to eliminate $\phi_{\underset{\sim}{k}}$ and $\psi_{\underset{\sim}{k}}$.

In the absence of an impurity $\rho_{\underset{\sim}{q}}(q{\neq}0) = 0$. In this case equation (2.28) reduces to the dispersion relation for a uniform electron gas

$$\omega^2_{(q)} = \omega^2_o \left[1 + \frac{9}{5} \frac{q^2}{k^2_o} \right] \quad . \tag{2.29}$$

Equation (2.29) agrees to order q^2 with the dispersion relation for a uniform electron gas obtained from a microscopic theory.[17]

We can compare our expression (2.28) with the corresponding equation obtained by Sziklas[11] and Sham[12] using a microscopic theory. In our notation their result can be written as

$$\frac{\omega^2}{\omega^2_o} \sigma_{\underset{\sim}{q}} = \frac{9}{5} \left(\frac{\omega_o}{\omega}\right)^2 \frac{q^2}{k^2_o} \sigma_{\underset{\sim}{q}} + \sigma_{\underset{\sim}{q}}$$

$$+ \sum_{\underset{\sim}{k'}\neq\underset{\sim}{q}} \frac{\underset{\sim}{q}\cdot\underset{\sim}{k'}}{k'^2} \frac{\rho_{\underset{\sim}{q}-\underset{\sim}{k'}}}{\rho_o} \sigma_{\underset{\sim}{k'}} \quad . \tag{2.30}$$

If we neglect the last two terms on the right hand side of equation (2.28) we obtain an equation which would be identical with equation (30) if the term $\frac{9}{5} (q^2/k^2_o) \sigma_{\underset{\sim}{q}}$

was replaced by

$$\frac{9}{5} \left(\frac{\omega_o^2}{\omega^2}\right) \frac{q^2}{k_o^2} \sigma_{\underset{\sim}{q}} \quad .$$

The factor ω_o^2/ω^2 does not account for the effect of the last two terms in equation (2.28), since this factor would be present in the case of a uniform electron gas, while the last two terms in equation (2.28) describe effects connected with the presence of the impurity. In equation (2.30) the only term describing the effect of the impurity on the density fluctuations (i.e. the third term on the R.H.S.) arises through the Coulomb interaction of the non-uniform (equilibrium) charge density with the potential due to the fluctuations. This can be seen by observing that the identical term in equation (2.28) arises from the second term on the right hand side of equation (2.26). The origin of the two extra terms in equation (2.28) is as follows: The first of these arises through the Coulomb interaction of the fluctuations with the potential produced by the non-uniform (equilibrium) electron charge density and by the fixed impurity. The second of these comes from the change in pressure due to fluctuations superimposed upon the non-uniform charge density. This interpretation follows from tracing the terms in equation (2.28) back to their origin in equation (2.26).

To solve equation (2.28) knowledge of $\rho_{\underset{\sim}{k}}$ is required. In the linearized Thomas-Fermi approximation

$$\rho_{\underset{\sim}{k}} = \frac{Z/\Omega}{1 + k^2/k_o^2} \qquad .$$

(2.31)

Substituting (2.31) into equation (2.28) we obtain

$$\left(\frac{\omega^2}{\omega_o^2} - 1 - \frac{9}{5}\frac{q^2}{k_o^2}\right)\sigma_{\underset{\sim}{q}} = \frac{z}{\Omega}\frac{1}{\rho_o} \sum_{\underset{\sim}{k}'\neq 0}\left[\frac{q\cdot k'[k_o^2/k'^2+1]+\frac{1}{5}q^2}{k_o^2 + |\underset{\sim}{q}-\underset{\sim}{k}'|^2}\right]\sigma_{\underset{\sim}{k}'} \quad .$$

(2.32)

We expand $\sigma_{\underset{\sim}{q}}$ in spherical harmonics, thus

$$\sigma_{\underset{\sim}{q}} = \sum_{\ell m}\sigma_q(\ell,m)\,Y_\ell^m(\theta_q,\,\phi_q) \quad .$$

(2.33)

Since we have assumed that the impurity is spherically symmetric $\sigma_q(\ell,m)$ does not depend on m. Thus we can write

$$\sigma_{\underset{\sim}{q}} = \sum_{\ell m}\sigma_q^\ell\,Y_\ell^m(\theta_q,\,\phi_q) \quad .$$

(2.34)

In terms of spherical harmonics equation (2.32) becomes

$$\left(\frac{\omega^2}{\omega_o^2} - 1 - \frac{9}{5}\frac{q^2}{k_o^2}\right)\sum_{\ell m}\sigma_q^\ell\,Y_\ell^m(\theta_q,\,\phi_q) = \frac{z}{(2\pi)^3}\frac{1}{\rho_o}$$

$$\times \int k'^2 dk'\sin\theta_{k'}\,d\theta\,d\phi_{k'}\left(\frac{qk'\,[k_o^2/k'^2+1]+\frac{1}{5}q^2}{k_o^2 + q^2 + k'^2 - 2qk'\mu}\right)$$

$$\times \sum_{\ell m}\sigma_{k'}^\ell\,Y_\ell^m(\theta_{k'},\,\phi_{k'})$$

(2.35)

where

$$\mu = \frac{\underset{\sim}{k'} \cdot \underset{\sim}{q}}{k'q} = \cos\theta_k \cos\theta_q + \sin\theta_{k'}\sin\theta_q\cos(\phi_{k'}-\phi_q).$$

Changing the variables of integration from $\theta_{k'}$, $\phi_{k'}$ to μ and ω, where ω is the azimuthal angle in the plane perpendicular to $\underset{\sim}{q}$ we obtain

$$(\frac{\omega^2}{\omega_o^2} - 1 - \frac{9}{5}\frac{q^2}{k_o^2}) \sum_{\ell m} \sigma_q^\ell \, Y_\ell^m(\theta_q,\phi_q) = \frac{Z}{(2\pi)^3}\frac{2\pi}{\rho_o} \int k'^2 dk' \int_{-1}^{1} du$$

$$\times \left[\frac{qk'\mu[k_o^2/k'^2+1] + \frac{1}{5}q^2}{k_o^2 + q^2 + k'^2 - 2qk'\mu}\right] \sum_{\ell m} \sigma_{k'}^\ell \, P_\ell(\mu) Y_\ell^m(\theta_q,\phi_q)$$

$$(2.36)$$

where we have used the relation

$$\int_0^{2\pi} Y_\ell^m(\theta_k,\phi_k)d\omega = 2\pi \, P_\ell^o(\mu) \, Y_\ell^m(\theta_q,\phi_q) \, . \qquad (2.37)$$

Making use of the orthogonality relations for the spherical harmonics equation (2.36) becomes

$$(\frac{\omega^2}{\omega_o^2} - 1 - \frac{9}{5}x^2) \, \sigma^\ell(x) = \frac{Z\,k_o^3}{4\pi^2\rho_o} \int_0^1 dx' \, B^\ell(x,x')dx' \qquad (2.38)$$

where $x = q/k_o$, $x' = k'/k_o$, and

$$B^{\ell}(x,x') = xx' \int_{-1}^{1} \frac{(1 + x'^2)\mu P_{\ell}(\mu) + \frac{1}{5} xx' P_{\ell}(\mu)}{1 + x^2 + x'^2 - 2xx'} du$$

$$= (1+x'^2)Q_1\left(\frac{1+x^2+x'^2}{2xx'}\right) + \frac{xx'}{5} Q_0\left(\frac{1+x^2+x'^2}{2xx'}\right) , \quad \ell=0$$

$$= \left[(1+x'^2)\left(\frac{x^2+x'^2+1}{2xx'}\right) + \frac{xx'}{5}\right]Q_{\ell}\left(\frac{1+x^2+x'^2}{2xx'}\right) , \quad \ell\neq0$$

$$(2.39)$$

Equation (2.38) can be solved numerically and the frequency spectrum obtained. We have restricted the range of integration in equation (2.38). This requires some comment. In the hydrodynamic model the integration should be over values of k from 0 to ∞. However the collective description of the electron gas breaks down for large values of $\underset{\sim}{k}$.[18] We have chosen the cut-off at $k=k_0$.[12] This is of the order of the maximum value of k for which collective modes are well defined in a uniform system.[12] Also one needs k's of the order of k_0 to "build" the distribution of charge in the neighbourhood of the impurity.

The argument of the Legendre functions of the second kind, Q_{ℓ}, which occur in equation (2.38), is always greater than or equal to 3/2, since

$$|\underset{\sim}{x}-\underset{\sim}{x}'|^2 \geq 0$$

implies

$$\frac{1 + x^2 + x'^2}{2xx'} \geq 1 + \frac{1}{2xx'}$$

and x and x' have a maximum value of unity.

Now $Q_\ell(y)$ for $y > 1$ is a rapidly decreasing function of ℓ, so that only the first few values of ℓ are important in equation (2.38). We have solved equation (2.38) numerically for $\ell = 0$ and $\ell = 1$. The results are given in figure (2.1). The result obtained by Sziklas is the same as (2.38) with $B^\ell(x,x')$ replaced by

$$B_s(x,x') = Q_1(\frac{1+x+x'^2}{2xx'}) \qquad\qquad \ell = 0$$

$$= (\frac{x^2+x'^2+1}{2xx'})Q_\ell(\frac{1+x+x'^2}{2xx'}) \qquad \ell \neq 0$$

Thus to the $\ell = 0$ mode we have a correction

$$x'^2Q_1 + \frac{xx'}{5} Q_o$$

and for the $\ell > 0$ mode a correction

$$(x'^2(\frac{x^2+x'^2+1}{2xx'}) + \frac{xx'}{5}) Q_\ell.$$

The localized modes exist above the continuum of "free" modes in the case of a positive impurity. The continuum of free modes extends from ω_o to $\sqrt{\frac{14}{5}} \omega_o$ (i.e. $q=k_o$ in (2.29)). From figure (2.1) we see that the localized mode has a frequency $\omega \sim \sqrt{3} \omega_o$.

Characteristic energy losses of k.e.v. electrons scattered from thin films have been attributed to plasma excitation. Comparison of results of experiments performed with pure materials and the same materials containing positive impurities should reveal the local frequencies predicted here. Even though the local frequencies lie close to the continuum they can be detected because they will absorb energy more readily since the impurity can take up momentum.
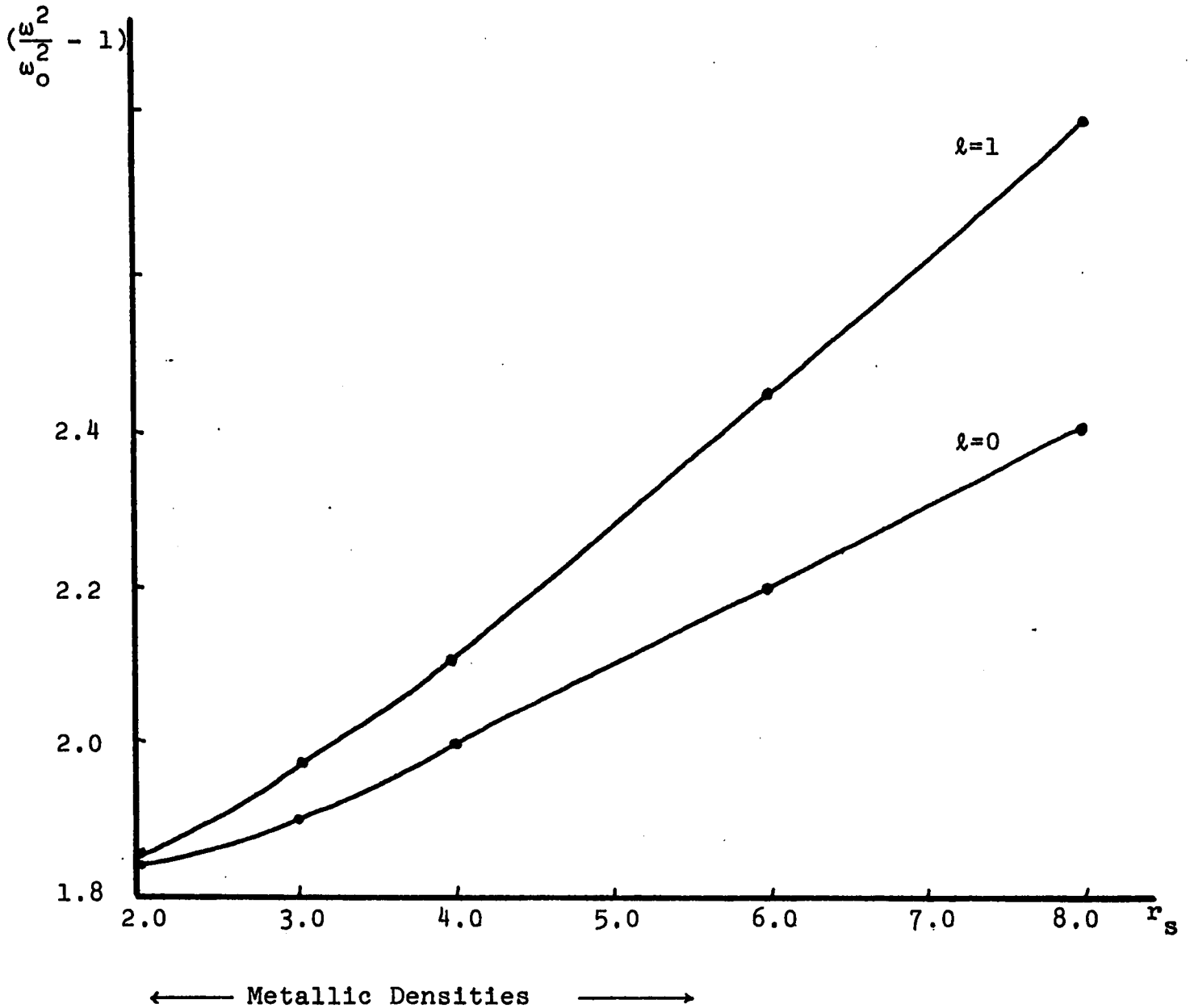
Figure II.1. Plot of $\omega^2/\omega_o^2 - 1$ vs r_s for $Z = +1$ and $\ell = 0; +1$, obtained numerically from eq.(2.38). Here r_s is defined by $\rho_o = \frac{4}{3} \pi (r_s a_o)^3$, where a_o is the Bohr radius.

PART II

INTRODUCTION

There exists a class of materials, called piezo-
electric, in which mechanical and electrical effects
are strongly interdependent. For instance if a piezo-
electric material is subject to a mechanical strain,
a macroscopic electric field appears in the crystal.
Conversely if the piezoelectric crystal is placed in
an electric field it becomes distorted.

Piezoelectric materials are insulators at room
temperature. However a piezoelectric crystal such as
CdS can be made semiconducting simply by shining light
on it. The electrons excited into the conduction band
in this manner will respond to an external electric field
and also to the macroscopic electric field associated
with the elastic vibrations of the crystal. The converse
effect will also occur, namely the electric field of the
conduction electrons will produce strains and hence vi-
brations in the crystal. Thus in a piezoelectric material
there is strong coupling between the electrons and the
lattice.

Besides the piezoelectric coupling the electrons
will interact with the optical polar modes (since piezo-
electric materials are ionic). In addition there will
be short range interactions between the electrons and
the lattice, i.e. deformation coupling. These

interactions may play an important role in determining
the transport properties of piezoelectric semiconductors,
but it is almost certain that it is the piezoelectric
coupling which is responsible for the most spectacular
experimentally observed effects.

Hutson et al.[23] studied the propagation of
acoustic waves in semiconducting CdS in the presence
of an applied D.C. field. They found that acoustic
waves of ~20 Mc are strongly amplified when the drift
velocity of the electrons exceeds the velocity of sound,
for waves travelling in the direction of the drift
velocity. Shortly after in 1962, Smith[24] observed
the phenomenon of current saturation in CdS. Upon
application of a high D.C. field the electric current
was observed to maintain a constant value for 10^{-7} sec
and then decay to a different constant value. The drift
velocity of the electrons at saturation was found to be
slightly greater than the longitudinal acoustic wave
velocity. Smith concluded that the saturation was a
direct consequence of the amplification of acoustic
waves. He postulated that energy and momentum were
transferred from the drifting electrons to travelling
acoustic waves.

In 1965 Yamashita and Nakamura[25] investigated
theoretically the phenomena of phonon amplification and
current saturation in piezoelectric semiconductors.

They used a quantum mechanical model in which the acoustic waves were quantized (phonons) and looked at three possible types of electron-phonon interaction, piezoelectric, optical and deformation. In the case of piezoelectric interaction they found that phonons of small wave number $q < 5 \times 10^5$ cm^{-1} are strongly amplified in a narrow cone around the direction of propagation of the drift current when the drift velocity exceeds a certain critical velocity, above the velocity of sound. They further found that as the number of phonons increases the current decreases to saturation in a time of approximately 10^{-6} sec. The calculations were done for CdS at room temperature with an electron number density 10^{15} cm^{-3}.

For the case of deformation and optical coupling they found that phonon amplification could not reasonably be expected to occur within their theory, at least at room temperature. This conclusion is in agreement with available experimental evidence.[26]

The theory developed by Yamashita and Nakamura can be applied for any value of the applied field. However there is a difficulty when it is applied to the region whose drift velocity exceeds the critical velocity which does not arise when the drift velocity is less than this critical value. Since in the former case the number of phonons is being increased enormously by the action of

the current, non-linear phonon-phonon interactions are
an extremely important process for establishing the
excited number of phonons in steady state. Indeed
without the non-linear phonon-phonon processes no steady
state is possible within their theory when the drift
velocity exceeds the velocity of sound. The inclusion
of non-linear processes in an exact fashion is a
formidable problem. Therefore Yamashita and Nakamura
introduced a cut-off in the phonon excitation number.
It was found that the time for saturation of the current
was not very sensitive to a choice of the value of the
cut-off, but that the saturation value of the drift
current was quite sensitive with regard to this value.
In Part II of this thesis we apply the theory of
Yamashita and Nakamura to the region where the drift
velocity is less than the critical velocity. In this
case the non-linear phonon-phonon processes play an un-
important role. However in this region excited phonons
may exist and influence the drift velocity. Their in-
fluence is quite different then in the case when the
drift velocity exceeds the critical velocity. In the
latter case the excess phonons reduce the current to
saturation value. When the drift velocity is less than
the critical velocity the effect of the induced phonons
is to increase the current above the value it would have
if the current were determined by the equilibrium dis-
tribution of phonon alone.

In Chapter I we develop the theory from a
different point of view than that of Yamashita and
Nakamura. We investigate the conditions for the
establishment of steady state of the electron-phonon
system in the presence of a D.C. field. In Chapter II
we apply the theory to the case of an A.C. electric
field applied to a piezoelectric semiconductor. In
Chapter III we discuss the results. We find that in
an A.C. field, below a critical value of the frequency
of the applied field, the magnitude of the drift
velocity of the electrons is increased over the value
it would have if it were determined by the equilibrium
distribution of phonons alone. This is due to the
effect of the induced phonons. Above this critical
frequency the non-equilibrium phonon distribution does
not have time to become established and the magnitude
of the drift velocity is reduced to a value determined
by the equilibrium distribution of phonons alone.

CHAPTER I

THEORY AND CONDITIONS FOR EXISTENCE

OF A STEADY STATE

I.1 Theory

We begin by considering the kinetic equation for

the electron distribution function $f_{\underset{\sim}{k}}$,

$$\frac{\partial f_{\underset{\sim}{k}}}{\partial t} = \frac{\partial f_{\underset{\sim}{k}}}{\partial t}\Bigg)_{e-p} + \frac{\partial f_{\underset{\sim}{k}}}{\partial t}\Bigg)_{Fi} \qquad (1.1)$$

where the suffixes e-p and Fi denote the terms arising

through the interaction of electrons with phonons and

the external field respectively. The kinetic equation

for the phonon distribution function $N_{\underset{\sim}{q}}$ is

$$\frac{\partial N_{\underset{\sim}{q}}}{\partial t} = \frac{\partial N_{\underset{\sim}{q}}}{\partial t}\Bigg)_{e-p} + \frac{\partial N_{\underset{\sim}{q}}}{\partial t}\Bigg)_{p-p} , \qquad (1.2)$$

where the suffixes e-p and p-p denote terms arising

through electron-phonon and phonon-phonon processes

respectively.

The relaxation time of an electron interacting

with lattice vibrations is of the order of 10^{-12} sec

at room temperature.   This is approximately the time

required for the electrons to establish a steady state

with the lattice. On the other hand the phonon-phonon relaxation time is of the order of 10^{-6} sec. Thus in the presence of an applied electric field the phonon distribution function will be a very slowly varying function of time compared to the electron distribution function. As long as the frequency of the applied field is much less than 10^{+12} sec, the electrons can be assumed to be in instantaneous steady state with the values of the external field and the number of phonons at any moment of time. Thus the left hand side of equation (1.1) is negligible for the frequency range of interest and we can write

$$\left.\frac{\partial f_{\underset{\sim}{k}}}{\partial t}\right)_{e-p} + \left.\frac{\partial f_{\underset{\sim}{k}}}{\partial t}\right)_{F1} = 0 \quad . \tag{1.3}$$

We wish to solve the coupled equations (1.2) and (1.3) in the presence of an electric field applied in the -z-direction. Ideally one would like to solve (1.2) and (1.3) for $f_{\underset{\sim}{k}}$ and $N_{\underset{\sim}{q}}$; in practice however this proves to be a hopelessly complicated task. We therefore use a method of partial solution developed by Fröhlich and Paranjape[27]. The idea is to guess a form of the solution for $f_{\underset{\sim}{k}}$ which contains a number of unknown parameters, and then use (1.2) and (1.3) to determine these parameters and $N_{\underset{\sim}{q}}$. It has been found[27] that if

the electron density is sufficiently high, then to a good approximation the electron distribution function $f_{\underset{\sim}{k}}$ is well represented by a displaced Maxwell Boltzmann distribution

$$f_{\underset{\sim}{k}} = (2\pi)^3 \, n_e \left(\frac{\hbar^2}{2\pi m k_B T}\right) \, e^{-\hbar^2 |\underset{\sim}{k} - \underset{\sim}{k}_o|^2 / 2mkT} \,, \qquad (1.4)$$

where n_e is the electron number density, m the electron mass, k_B Boltzmann constant. Mathematically, k_o and T are two unknown parameters which are to be determined. Physically, T is interpreted as the electron temperature, not necessarily equal to the lattice temperature T_o and $\hbar k_o/m$ as the electron drift velocity v_d. The two parameters k_o and T are determined by requiring that the rate of transfer of energy and momentum by the field to the electrons is equal to that transferred from the electrons to the lattice. Mathematically this corresponds to taking the first two moments of equation (1.3), which illustrates the approximation nature of the solution. We follow Yamashita and Nakamura in assuming that $T=T_o$. Thus we only use one of the above mentioned conditions, namely that the rate of transfer of momentum from the field to the electrons is equal to that transferred from the electrons to the lattice.

The rate of transfer of momentum from the field to the electrons is

$$\sum_{\underset{\sim}{k}} \hbar k \left.\frac{\partial f_k}{\partial t}\right)_{F1} = e F N \quad , \qquad (1.5)$$

where e is the magnitude of the charge on the electron, N is the total number of electrons and F is the applied field in the -z direction. Equation (1.5) is just Newton second law. The rate of transfer of momentum from the electrons to the lattice is (neglecting U-processes)

$$\sum_{\underset{\sim}{k}} \hbar k \left.\frac{\partial f_k}{\partial t}\right)_{e-p} = \sum_{\underset{\sim}{q}} \hbar q \left.\frac{\partial N_q}{\partial t}\right)_{e-p} = e F N \quad . \qquad (1.6)$$

We will mainly be concerned with the effects of an A.C. electric field on a material in which the electron-phonon interaction is exclusively piezoelectric. For the moment however we will include the effects of polar interactions, so that we can compare our resulting equations with those obtained by Yamashita and Nakamura. Polar interactions are important in many piezoelectric semiconductors at room temperature. In fact in CdS at room temperature the interaction of the electrons with the polar modes is mainly responsible for determining the low field mobility.[25] However the polar interactions do not give rise to an appreciable number of non-equilibrium phonons (because of large energy required to excite them) and we assume that their effect may be adequately

described by the introduction of a relaxation time τ_0. Hence we write

$$\sum_{\underset{\sim}{k}} \hbar \underset{\sim}{k} \left. \frac{\partial f_{\underset{\sim}{k}}}{\partial t} \right)_{e-p} = \sum_{\underset{\sim}{k}} \hbar \underset{\sim}{k} \left. \frac{\partial f_{\underset{\sim}{k}}}{\partial t} \right)_{polar} + \sum_{\underset{\sim}{k}} \hbar \underset{\sim}{k} \left. \frac{\partial f_{\underset{\sim}{k}}}{\partial t} \right)_{p.e.}$$

$$= - \sum_{\underset{\sim}{k}} \hbar \underset{\sim}{k} \left(\frac{f_{\underset{\sim}{k}} - f_{\underset{\sim}{k}}^0}{\tau_0} \right) + \sum_{\underset{\sim}{q}} \hbar \underset{\sim}{q} \left. \frac{\partial N_{\underset{\sim}{q}}}{\partial t} \right)_{p.e.} \qquad (1.7)$$

where

$$\left. \frac{\partial f_{\underset{\sim}{k}}}{\partial t} \right)_{polar} = - \frac{f_{\underset{\sim}{k}} - f_{\underset{\sim}{k}}^0}{\tau_0} \quad,$$

$f_{\underset{\sim}{k}}^0$ is the equilibrium distribution, and p.e. stands for piezoelectric. Thus from (1.7), (1.6) and (1.2) we have the following two coupled equations

$$- \sum_{\underset{\sim}{k}} \hbar \underset{\sim}{k} \frac{f_{\underset{\sim}{k}} - f_{\underset{\sim}{k}}^0}{\tau_0} + \sum_{\underset{\sim}{q}} \hbar \underset{\sim}{q} \left. \frac{\partial N_{\underset{\sim}{q}}}{\partial t} \right)_{p.e.} = e F N \qquad (1.8a)$$

$$\frac{\partial N_{\underset{\sim}{q}}}{\partial t} = \left. \frac{\partial N_{\underset{\sim}{q}}}{\partial t} \right)_{p.e.} + \left. \frac{\partial N_{\underset{\sim}{q}}}{\partial t} \right)_{p-p} \qquad \cdot \qquad (1.8b)$$

Making use of (1.4), equations (1.8) are to be solved for the electron drift velocity v_d and $N_{\underset{\sim}{q}}$.

The term

$$\left. \frac{\partial N_{\underset{\sim}{q}}}{\partial t} \right)_{p.e.}$$

which appears in equations (1.8a) and (1.8b) can be evaluated using Fermi's Golden Rule. The probability per unit time for an electron in a plane wave state $k+q$ to make a transition to a state k with emission of a phonon q is

$$\frac{2\pi}{\hbar} c_q^2 \, f_{k+q}(N_q+1) \, \delta(\varepsilon_{k+q} - \varepsilon_k - \hbar\omega q) \, .$$

c_q^2 is the square of the matrix element of the piezo-electric interaction labelled by $k+q$ and k; $\hbar\omega q$ is the energy of the emitted phonon; $\varepsilon_k = \hbar^2 k^2/2m$.

Similarly the rate of transition of an electron from a plane wave state k to a state $k+q$ with the absorption of a phonon of wave vector q is

$$\frac{2\pi}{\hbar^2} c_q^2 f_k N_q \, \delta(\varepsilon_{k+q} - \varepsilon_k - \hbar\omega q) \, .$$

Therefore

$$\left.\frac{\partial N_q}{\partial t}\right)_{p.e.} = \sum_k \frac{2\pi}{\hbar^2} c_q^2 \{ f_{k+q}(N_q + 1) - f_k N_q \} \delta(\varepsilon_{k+q} - \varepsilon_k - \hbar\omega q) \, . \tag{1.9}$$

We expand the displaced Maxwell Boltzmann distribution, equation (1.4), as follows:

$$f_k \approx f_o(\varepsilon_k) - \hbar v_d kx \frac{\partial f_o}{\partial \varepsilon} \, . \tag{1.10}$$

Similarly

$$f_{k+q} = f_o(\epsilon_{k+q}) - \hbar v_d(k_x + q_x) \frac{\partial f_o}{\partial \epsilon} \quad . \qquad (1.11)$$

The collision of the electron with the lattice is nearly elastic owing to the relatively small mass of the electron. Hence we expand $f_o(\epsilon_{k+q})$ as follows:

$$f_o(\epsilon_{k+q}) \approx f_o(\epsilon_k) + \hbar \omega q \frac{\partial f_o}{\partial \epsilon} \quad . \qquad (1.12)$$

Using the approximations (1.10), (1.11) and (1.12) equation (1.9) can be evaluated using standard methods.[28] We give the final result

$$\left. \frac{\partial \xi_q}{\partial t} \right)_{p.e.} = \frac{V c_q^2}{2\pi} \left(\frac{m}{\hbar^2}\right)^2 f_o(\epsilon_q)(v_d \cos\theta - s)\, \xi_q$$

$$+ \frac{V c_q^2}{2\pi} \left(\frac{m}{\hbar^2}\right)^2 f_o(\epsilon_q)\, v_d \cos\theta\, n_q^{o*} \qquad (1.13)$$

where V is the volume of the sample;

$\quad \theta$ is the angle between q and the applied field;

$\quad \xi_q = N_q - n_q^o$, where n_q^o is the Planck distribution;

$\quad \omega_q = s\, q$, where s is the velocity of sound;

and $\quad n_q^{o*} = n_q^o + \frac{1}{2}$.

For a piezoelectric interaction[29]:

$$c_q^2 = \frac{1}{V} \left(\frac{4\pi\beta e}{\epsilon_o}\right)^2 \frac{\hbar}{2MN_a s} \left[\frac{q^2}{q^2 + q_o^2}\right]^2 \frac{1}{q} \quad , \qquad (1.14)$$

where β = piezoelectric constant;

ε_o = dielectric constant;

N_a = no. of atoms/cm^3;

q_o = $(4\pi n_e^2/\varepsilon_o kT)$ is the inverse Debye screening length;

M = mass of an atom.

In equation (1.13) we make the substitutions

$$x = \frac{\hbar q}{\sqrt{2mk_BT}} \quad , \quad y = \cos\theta \quad\quad (1.15)$$

and rewrite (1.13) as

$$\left.\frac{\partial\xi(x,y)}{\partial t}\right|_{p.e.} = \frac{g(x)}{\ell}\{(v_dy-s)\xi(x,y) + v_dyn_o^*\} \ , \quad\quad (1.16)$$

where

$$g(x) =(2\pi)^3\, n_e\left(\frac{\hbar^2}{2mk_BT}\right)^{3/2}\frac{1}{x}\left(\frac{x^2}{x^2+x_o^2}\right)e^{-\frac{1}{4}x^2}$$

$$\frac{1}{\ell} = \frac{1}{2\pi}\left(\frac{4\pi\beta e}{\varepsilon_o}\right)^2\frac{1}{2MN_a s\hbar^2}\ \frac{1}{\sqrt{2mkT}}\ m^2 \ .$$

Substituting (1.16) into (1.8a) we obtain

$$eF_{ne} = \frac{m\,v_d\,n_e}{\tau_o} + \hbar\left(\frac{2mkT}{\hbar^2}\right)^2\frac{1}{\ell}\frac{1}{4\pi^2}\int dx\ x^3 g(x)$$

$$\int dy\{(v_dy^2 - sy)\xi(x,y) + v_dy^2n_o^*\} \ . \quad\quad (1.17)$$

Solving (1.17) for v_d we obtain

$$v_d = \frac{\frac{eF}{m} + Bs \int h(x)dx \int y\, \xi(x,y)\, dy}{\frac{1}{\tau_o} + \frac{1}{\tau_{p.e.}} + B\int h(x)dx \int y^2 dy\, \xi(x,y)} \quad , \qquad (1.18)$$

where

$$B = \frac{1}{\ell}\, 2\, \sqrt{\frac{2k_B T}{m}}$$

$$h(x) = x^2\left(\frac{x^2}{x^2 + x_o^2}\right)^2 e^{-\frac{1}{4}x^2}$$

and

$$\frac{1}{\tau_{p.e.}} = B \int h(x)\, dx \int y^2\, dy\, n_o^* \quad . \qquad (1.18a)$$

At room temperature the low field mobility in CdS is determined mainly by the polar interactions, in which case $\frac{1}{\tau_o} \gg \frac{1}{\tau_{p.e.}}$. If we neglect $1/\tau_{p.e.}$ compared with $1/\tau_o$ in (1.18) we obtain an equation which is identical with that obtained by Yamashita and Nakamura. However we are interested in the case where $1/\tau_o \rightarrow 0$ i.e. pure piezoelectric coupling and we write equation (1.18) as

$$v_d = \frac{\mu_{p.e.}\, F + B\tau_{p.e.} \int h(x)dx \int y\xi(x,y)dy}{1 + B\tau_{p.e.} \int h(x)dx \int y^2 \xi(x,y)dy} \quad , \qquad (1.19)$$

where

$$\mu_{p.e.} = \frac{e\tau_{p.e.}}{m} \quad .$$

The effect of non-equilibrium phonons on the drift velocity is contained in $\xi(x,y)$ in equation (1.19). If the effect of the non-equilibrium phonons is small then

$$v_d \approx \mu_{p.e.} F \; .$$

(1.20)

An expression for $\xi(x,y)$ is obtained from equation (1.8b). To solve this equation we need information on $\partial N_{\underset{\sim}{q}}/\partial t]_{p-p}$. As long as the drift velocity is less than the velocity of sound we follow Yamashita and Nakamura in assuming that the phonon-phonon interaction is adequately described by a phonon relaxation time, i.e.

$$\frac{\partial N_{\underset{\sim}{q}}}{\partial t}\Bigg)_{p-p} = - \frac{N_{\underset{\sim}{q}} - n^o_{\underset{\sim}{q}}}{\bar{\tau}_{\underset{\sim}{q}}} = - \frac{\xi_{\underset{\sim}{q}}}{\bar{\tau}_{\underset{\sim}{q}}} \; .$$

(1.21)

We can then write equation (1.8b), using equations (1.16) and (1.21) as

$$\frac{\partial \xi(x,y,t)}{\partial t} = \frac{g(x)}{\ell} \{v_d y - s^*\}\xi(x,y,t) + \frac{g(x)}{\ell} v_d y \; n^*_o$$

(1.22)

where

$$s^* = s + \frac{\ell}{\bar{\tau}(x)g(x)} \; .$$

(1.23)

The two coupled equations (1.18) and (1.22) can be solved and a time dependent solution for the drift velocity due to a time dependent electric field obtained.

It is of interest however to obtain the steady state solution of (1.18) and (1.22) when $v_d < s^*$.

I.2 Steady State

We first note that equation (1.22) has no steady state solution for $v_d y > s^*$. In this case it is necessary to include non-linear phonon-phonon interactions in order to ensure the existence of a steady state. However when $v_d < s^*$ equation (1.22) has the physically acceptable steady state solution

$$\xi(x,y) = \frac{v_d y\ n_o^*(x)}{s^*(x) - v_d y} \quad . \tag{1.24}$$

We substitute equation (1.24) into equation (1.18). The integrations over y are straightforward and are given by

$$\int_{-1}^{+1} y\xi(x,y)dy = -\frac{2s^*}{v_o} + \frac{s^{*2}}{v_o^2} \ln\left|\frac{1+v_o/s^*}{1-v_o/s^*}\right| \tag{1.25}$$

and

$$\int_{-1}^{+1} y^2\xi(x,y)dy = -\frac{2}{3} - \frac{2s^{*2}}{v_o^2} - \frac{s^{*3}}{v_o^3} \ln\left|\frac{1+v_o/s^*}{1-v_o/s^*}\right| \tag{1.26}$$

where v_o is the steady state value of the drift velocity.

Substituting (1.25) and (1.26) into (1.18) we obtain

$$v_o = \frac{\mu_{p.e.}F_o + Bs_{p.e.}\int h(x)dx\left[-\frac{2s^*(x)}{v_o} + \frac{s^{*2}(x)}{v_o^2}\ln\left|\frac{1+v_o/s^*(x)}{1-v_o/s^*(x)}\right|\right]}{1 + B\tau_{p.e.}\int h(x)dx\left[-\frac{2}{3}\frac{2s^{*2}(x)}{v_o^2} - \frac{s^*(x)}{v_o^3}\ln\left|\frac{1+v_o/s^*(x)}{1-v_o/s^*(x)}\right|\right]}$$

(1.27)

Multiplying both sides of equation (1.27) by the denominator on the left hand side, expanding the logarithms $(v_o/s^* < 1)$ and collecting powers of v_o, we obtain

$$v_o[1 - B\tau_{p.e.}\int\frac{2}{3}n_o^*h(x)\,dx\,\frac{s}{s^*}] + B\tau\int\frac{2}{3}h(x)dx\,n_o^*(1-\frac{s}{s^*})$$

$$\times\left[\frac{3}{5}\frac{v_o^5}{s^{*5}} + \frac{3}{7}\frac{v_o^7}{s^{*7}} + \frac{3}{9}\frac{v_o^9}{s^{*9}} + \dots\right]\frac{s^{*3}}{v_o^2} = \mu_{p.e.}F_o \quad .$$

(1.28)

The sum in the curly bracket extends to infinity. Remembering the definition of $\tau_{p.e.}$, and expressing the power series in v_o as a logarithm, equation (1.28) can be written as

$$B\tau_{p.e.}\int\frac{2}{3}h(x)n^o(x)(1-s/s^*(x))\frac{s^{*3}(x)}{v_o^2}$$

$$\times\left[\frac{3}{2}\ln\left|\frac{1+v_o/s^*}{1-v_o/s^*}\right| - \frac{3v_o}{s^*}\right]dx = \mu_{p.e.}F_o \quad . \quad (1.29)$$

Equation (1.29) is exact and is an expression for the steady state drift velocity v_o in terms of the

applied D.C. field F_o. If we consider the case when $v_o/s^*(x) \ll 1$, then expanding equation (1.29) in powers of v_o/s^* and keeping terms linear in v_o/s^* we obtain

$$v_o B \tau_{p.e.} \int \frac{2}{3} h(x) n_o^*(1 - s/s^*(x)) dx = \mu_{p.e.} F_o . \qquad (1.30)$$

From equation (1.30) the low field D.C. mobility μ is

$$\mu = \frac{\mu_{p.e.}}{B \tau_{p.e.} \int \frac{2}{3} h(x) n_o^*(1 - s/s^*(x)) dx} . \qquad (1.31)$$

If an experiment is performed to measure the low field mobility, one presumably measures (1.31). If we look at the definition of $s^*(x)$, i.e. equation (1.23) we see that if $\bar{\tau}(x)$ is very small then $s/s^*(x) \sim 0$. In this case it follows from equation (1.31) that $\mu \approx \mu_{p.e.}$. However we will find that in a reasonable model of a physical material CdS, $\bar{\tau}(x)$ is sufficiently long so that μ is appreciably different from $\mu_{p.e.}$.

It follows from equation (1.30) and also from the exact equation (1.29) that if s/s^* is sufficiently close to unity then no steady state solution exists for $v_o < s$. The fact that no steady state solution exists when $s/s^* \sim 1$ is not apparent from the equation for the non-equilibrium phonons. In fact replacing $s^*(x)$ by s in equations (1.22) or (1.24) seems to make no difference as far as the formal existence of a steady state is

concerned. But the existence of a steady state $\xi(x,y)$ depends on the existence of a steady state drift velocity for which it is necessary that $s \neq s^*$.

From equation (1.24) we see that the effect of the current is to increase the number of phonons moving in the direction of the current ($y > 0$) and decrease the number moving in the opposite direction ($y < 0$). The result of this is to increase the electron mobility, μ, above the value it would have if determined by the equilibrium distribution of phonon alone, i.e. $\mu_{p.e.}$. To discover whether this effect is important one must have some method of measuring $\mu_{p.e.}$. One way of measuring $\mu_{p.e.}$ would be to apply an electric field for a very short time, since the non-equilibrium distribution of phonons requires a finite time to establish itself. If the applied "pulse" was of short enough duration, the mobility of the electrons would be determined exclusively by the equilibrium distribution of phonons. What would seem to be a more practical method would be to apply an A.C. field of high enough frequency. In this case the current would never be in one direction long enough to excite phonons.

CHAPTER II

APPLICATION TO A.C. FIELD

We consider a pure A.C. field of the form

$$F(t) = F \sin \omega t \quad .$$ (2.1)

This field gives rise to a time dependent drift current, which in turn gives rise to a time dependent phonon distribution. If the frequency ω is small enough, then at a given instant the electrons and phonons are almost in steady state, and the phonon distribution is to a good approximation given by equation (1.24). However above some frequency of the applied field the phonons will never reach steady state, and equation (1.24) is no longer applicable. At even higher frequencies we expect on the basis of the intuitive argument given at the end of Chapter I that non-equilibrium phonons are not present in appreciable numbers at all. The time required for the electron phonon system to reach steady state for a constant drift velocity can be obtained from equation (1.22), and is of the order

$$- \frac{\ell}{g(x)\{v_d y - s^*\}} \sim - 10^{-3} \frac{1}{(v_d y - s^*)}$$ (2.2)

in CdS at $T = 30^\circ K$, $n_e = 10^{15} \ cm^{-3}$. At low drift velocity this time is of the order of 10^{-8} sec.

We first considered the general form of the
phonon distribution function. If the current is moving
in the +z direction it emits phonons in that direction.
When the direction of the current is reversed it begins
to emit phonons in the -z direction. Since the phonons
have a finite lifetime $\bar{\tau}$ the phonons emitted in the +z
direction will not have vanished when the current is
reversed. Hence at sufficiently high frequencies but
not too high there are always some induced phonons
present. The phonon distribution function therefore
does not oscillate in time around its equilibrium value,
but about some value in excess of this. We express this
fact by writing

$$\xi(x,y,t) = \xi^o(x,y) + \xi^1(x,y,t) \qquad (2.3)$$

where ξ^o is time independent. Provided the material has
the same properties in the +z and -z directions, $\xi^o(x,y)$
must be an even function of y. Thus

$$\xi^o(x,y) = \xi^o(x,-y) . \qquad (2.4)$$

Substituting (2.3) into (1.22), and equating the time
independent parts we obtain

$$\xi^o(x,y)s^* = \text{Time independent part of } (g(x)v_d(t)\xi^1(x,y,t)y).$$

Since $\xi^o(x,y)$ is an even function of y, provided s^* is

independent of y, which we assume is the case, $\xi^1(x,y)$ is an odd function of y and hence

$$\int_{-1}^{1} y\xi^0 dy = \int_{-1}^{1} y^2 \xi^1 dy = 0 . \qquad (2.5)$$

Substituting (2.1) and (2.3) into (1.19) and making use of (2.5) we have

$$v_d(t) = \frac{\mu_{p.e.} F(t) + Bs\tau_{p.e.} \int h(x)dx \int \xi^1(x,y,t)y\,dy}{1 + B\tau_{p.e.} \int h(x)dx \int y^2 \xi^0(x,y)dy} . \qquad (2.6)$$

Only the numerator of the right hand side of (2.6) is time dependent. If $v_d(t)$ and $\xi^1(x,y,t)$ are expanded in a Fourier series in time then the lowest harmonic of $v_d(t)$ (i.e. the one that oscillates with frequency ω) will be out of phase with both the applied field and the lowest harmonic of $\xi^1(x,y,t)$. All higher harmonics of the current will be in phase with the corresponding harmonics of $\xi^1(x,y,t)$. In other words, all harmonics of the current higher than the first are produced exclusively by the action of the non-equilibrium phonons. We neglect this effect and concentrate on the first harmonic of the current which is produced by both the action of the field and the non-equilibrium phonons.

We look for a response to the applied field which has the form

$$v_d(t) = v_1 \sin(\omega t + \delta_1) \tag{2.7}$$

$$\xi(x,y,t) = \xi^0(x,y) + \xi^1(x,y)\sin(\omega t + \Delta_1(x)) . \tag{2.8}$$

Equations (2.7) and (2.8) contain 5 unknowns, v_1, δ_1, $\xi^0(x,y)$, $\xi^1(x,y)$ and $\Delta_1(x)$. Substituting (2.7) and (2.8) into (1.22) and equating the linearly independent terms (i.e. the coefficients of 1, $\sin \omega t$ and $\cos \omega t$) we obtain the following three relations.

$$\xi^0(x,y) = -n_o^* + \cfrac{n_o^*}{1 - \cfrac{(v_1 y)^2}{2}\left(\cfrac{g(x)}{\ell}\right)^2\left[\omega^2 + \left[\cfrac{g(x)s^*(x)}{\ell}\right]^2\right]^{-1}} \tag{2.9}$$

$$\xi^1(x,y) = \cfrac{v_1 y\, g(x)}{\ell\sqrt{\omega^2 + \left[\cfrac{g(x)s^*(x)}{\ell}\right]^2}}\ \cfrac{n_o^*}{1 - \cfrac{(v_1 y)^2}{2}\left(\cfrac{g(x)}{\ell}\right)^2\left\{\omega^2 + \left(\cfrac{g(x)s^*(x)}{\ell}\right)\right\}^{-1}} \tag{2.10}$$

and

$$\delta_1 = \tan^{-1}\frac{\omega\ell}{g(x)s^*(x)} + \Delta_1(x) . \tag{2.11}$$

Equations (2.9) and (2.10) give ξ^0 and ξ^1 as functions of one unknown v_1 and equation (2.11) relates δ_1 and $\Delta_1(x)$. Substituting (2.1), (2.7) and (2.9) into (2.6) and equating linearly independent terms we obtain

$$v_1 \cos \delta_1 = -\mu_{p.e.} \frac{F+sB_{p.e.} \int h(x)dx \int y\xi^1(x,y)\{\gamma(x)s^* \cos\delta_1 + \frac{\omega\ell\gamma(x)}{g(x)}\sin\delta_1\}dy}{1 + B\tau_{p.e.} \int h(x)dx \int y^2 dy \xi^0(x,y)}$$

$$(2.12)$$

$$v_1 \sin\delta_1 = \frac{sB\tau_{p.e.} \int h(x)dx \int y\xi^1(x,y)dy\{\gamma(x)s^* \sin\delta_1 - \frac{\omega\ell\gamma(x)}{g(x)}\cos\delta_1\}}{1 + B\tau_{p.e.} \int h(x)dx \int y^2 dy \xi^0(x,y)}$$

$$(2.13)$$

where

$$\gamma(x) \equiv \frac{g(x)/\ell}{\omega^2 + \left(\frac{g(x)s^*}{\ell}\right)^2} \qquad . \qquad (2.14)$$

Equations (2.12) and (2.13) can be solved for v_1 and δ_1 when ξ^0 and ξ^1 are substituted from (2.9) and (2.10).

We have assumed that s^* is not a function of y. In this case the integrations over y in (2.12) and (2.13) can be performed in a straightforward manner. The results are

$$\int_{-1}^{+1} y^2 \xi^0(x,y)dy = n_o^* \left[-\frac{2}{3} + \frac{1}{v_1^2\gamma^2}\left(-4 + \frac{2\sqrt{2}}{v_1\gamma} \log \left| \frac{1 + \frac{v_1\gamma}{\sqrt{2}}}{1 - \frac{v_1\gamma}{\sqrt{2}}} \right| \right) \right]$$

$$(2.15)$$

and

$$\int_{-1}^{1} y\xi^1(x,y)dy = \frac{n_o^*}{v_1\gamma}\left(-4 + \frac{2\sqrt{2}}{v_1\gamma} \log\left| \frac{1 + \frac{v_1\gamma}{\sqrt{2}}}{1 - \frac{v_1\gamma}{\sqrt{2}}} \right| \right) \qquad . \qquad (2.16)$$

We substitute (2.15) and (2.16) into (2.12) and (2.13) and multiply both sides of these equations by the denominators occurring on the right hand sides of (2.12) and (2.13). Collecting the coefficients of $\cos\delta_1$ and $\sin\delta_1$ we note that they contain terms involving

$$\ln \frac{1 + v_1/\sqrt{2}}{1 - v_1/\sqrt{2}} \quad .$$

These coefficients can be regrouped in a neat form if one expands the logarithms $((\gamma v_1/\sqrt{2}) < 1)$, and collects powers of v_1. This power series can be again expressed in terms of a logarithm. This procedure is analogous to the method by which equation (1.29) was obtained from equation (1.27). We then obtain the following two equations :

$$\cos\delta_1 \left[B_{p.e.} \int h(x)n_0^* \{1 - \gamma^2 ss^*\}\frac{2\sqrt{2}}{v_1^2\gamma^3} \{ \ln \left| \frac{1 + \frac{v_1\gamma}{\sqrt{2}}}{1 - \frac{v_1\gamma}{\sqrt{2}}} \right| \right.$$

$$- \frac{2v_1\gamma}{\sqrt{2}}\} dx = \mu_{p.e.}F + \sin\delta_1 \{B\tau_{p.e.} \int [h(x)n_0^*$$

$$\times \frac{\omega\ell}{g(x)} \frac{s2\sqrt{2}}{v_1^2\gamma^2} \{ \ln \left| \frac{1 + \frac{v_1\gamma}{\sqrt{2}}}{1 - \frac{v_1\gamma}{\sqrt{2}}} \right| - \frac{2v_1\gamma}{\sqrt{2}}\}]dx \right) \qquad (2.17)$$

$$\tan\delta_1 = -\frac{\int h(x)\ n_o^*\ \frac{\omega \ell s}{g(x)}\ \frac{1}{\gamma}\left[\ln\left|\frac{1 + \frac{v_1\gamma}{\sqrt{2}}}{1 - \frac{v_1\gamma}{\sqrt{2}}}\right| - \frac{2v_1\gamma}{\sqrt{2}}\right]\ dx}{\int h(x)\ n_o^*(1 - \gamma^2 ss^*)\ \frac{1}{\gamma^3}\left[\ln\left|\frac{1 + \frac{v_1\gamma}{\sqrt{2}}}{1 - \frac{v_1\gamma}{\sqrt{2}}}\right| - \frac{2v_1\gamma}{\sqrt{2}}\right]\ dx}$$

$$(2.18)$$

CHAPTER III

DISCUSSION .

III.1 The phonon distribution function

We give some arguments to justify the form of the equations obtained in Chapter II. The decomposition of $\xi(x,y,t)$ (2.8) was arrived at on the basis that at high enough frequencies there are always some non-equilibrium phonons present and therefore N_q does not oscillate around its equilibrium value but at some value in excess of this. One might therefore expect that $\xi^o(x,y)$ should tend to zero at low frequencies. However that this is not the case can be seen by the following arguments. When the current is moving in a certain direction it depletes below their equilibrium value the number of phonons travelling in the opposite direction. However, it cannot deplete this number by any move than the number present in equilibrium, i.e. n_q^o. There is no such restriction on the number of phonons the current can create in the forward direction. In steady state the number of phonons created in the forward direction is from equation (1.24)

$$\frac{v_d y \; n_o^*(x)}{s^* - v_d y} \quad .$$

The depletion in the backward direction is

$$- \frac{v_d y n_o^*}{s^* + v_d y} \quad .$$

Hence the average number of non-equilibrium phonons in the two modes (x,y), $(x,-y)$ is

$$\xi_{av}(x,y,-y) = (\frac{v_d y}{s^*})^2 \ \frac{n_o^*}{1 - (\frac{v_d y}{s^*})^2} \quad . \tag{3.1}$$

In the A.C. case the two modes (x,y) and $(x,-y)$ are treated together, as can be seen if we recall that the angle y is always measured from the direction of the current, and that the current changes direction over a cycle of the applied field. Thus when the current is moving in the +z direction it is increasing the number of phonons in the mode (x,y) where y is measured from +z direction. When the current changes direction the mode formerly (x,y) becomes $(x,-y)$ measured from the -z direction. Thus at low frequency we expect $\xi(x,y,t)$ to oscillate above its average value given by (3.1). During a cycle v_d^2 varies between 0 and v_1^2 where v_1 is the maximum value of the drift velocity. Replacing v_d^2 by its average value over a cycle, i.e. $v_1^2/2$, we obtain from (3.1)

$$\lim_{\omega \to 0} \xi^{o}(x,y) = \left(\frac{v_1 y}{\sqrt{2}\, s^*}\right)^2 \frac{n_o^*}{1 - \left(\frac{v_1 y}{\sqrt{2}\, s^*}\right)^2} \qquad . \qquad (3.2)$$

Taking the limit of equation (2.9) as $\omega \to 0$ gives equation (3.2).

Let us subtract out from the steady state, equation (1.24) the average value (3.1)

$$\xi - \xi_{av} = \frac{v_d y}{s^*} \frac{n_o^*}{1 - \left(\frac{v_d y}{s^*}\right)^2} \qquad . \qquad (3.3)$$

Thus at low frequencies ξ should oscillate about its average value ξ_o with an amplitude given by (3.3). Thus

$$\lim_{\omega \to 0} \xi^{1}(x,y) = \frac{v_1 y}{s^*} \frac{n_o^*}{1 - \left(\frac{v_1 y}{\sqrt{2}\, s^*}\right)^2} \qquad (3.4)$$

where v_1 has been substituted for v_d in the numerator since the maximum value of the drift velocity determines the maximum value of ξ^1. Taking the limit of equation (2.10) as $\omega \to 0$ gives (3.4).

Having given a meaning to the expression

$$\xi(x,y,t) = \xi_o + \xi_1 \sin \omega t \qquad . \qquad (3.5)$$

At low frequencies, it is necessary to point out that equation (3.5) does not constitute a good physical

description at low frequencies, i.e. $\omega \ll 1/\bar{\tau}$.

Equation (3.5) implies that as the current goes to

zero during a cycle, i.e. at times $t = 2\pi n/\omega$, the

non-equilibrium phonon represented by ξ^0 will be

present. Since ξ^0 is the time averaged value of the

number of phonons in a mode, this will only be the

case when $\omega \gg 1/\bar{\tau}$. In CdS at $40°K$, $1/\bar{\tau} \sim 10^5$ sec^{-1}.

III.2 Effect of the phonon distribution on the current

To obtain a rough idea of how the solutions of

(2.17) and (2.18) are expected to behave, we assume

that $s^*(x)$, $\gamma(x)$ and $g(x)$ may be taken out from under

the integral sign, and replaced by their values at the

maximum value of the integrand, which we denote by $\bar{s}^*$,

$\bar{\gamma}$ and $\bar{g}$. Then from (2.18) we obtain

$$\tan\delta_1 = -\frac{\dfrac{\omega\ell}{\bar{g}}\dfrac{s}{s^{*2}}}{\{\dfrac{\omega^2\ell}{\bar{g}^2\bar{s}^{*2}} + (1 - \dfrac{s}{\bar{s}^*})^2\}} \qquad . \tag{3.6}$$

From (2.17) we obtain, to terms linear in v_1

$$v_1 = \frac{\mu_{p.e.}F\{\dfrac{\omega^2\ell^2}{\bar{g}^2\bar{s}^{*2}} + 1\}^{\frac{1}{2}}}{\{\dfrac{\omega^2\ell^2}{\bar{g}^2\bar{s}^{*2}} + (1 - s/\bar{s}^*)^2\}^{\frac{1}{2}}} \qquad . \tag{3.7}$$

In (3.7) as ω becomes very large v_1 approaches $\mu_{p.e.}F$. At smaller values of ω the effect of the phonons is to increase the value of v_1 above $\mu_{p.e.}F$. The high frequency limit is exact and follows from (2.17) and (2.18).

Equations (2.17) and (2.18) were solved numerically for CdS at $T = 30^\circ K$ and $n_e = 10^{15}$. At temperatures below $50^\circ K$ the coupling between the lattice and the electrons is piezoelectric.[30] Above this temperature polar coupling becomes important. An expression for $\bar{\tau}(x)$ at the temperature we are interested in has been given by Holland[31]

$$\frac{1}{\bar{\tau}_q} = D\, T^3 s^2 q^2 \qquad (3.8)$$

where $D = 2.74 \times 10^{-22}$ sec/T^3. However this value of D gives the phonons a mean free path many times larger than the average size of experimental sample. We have therefore chosen

$$\frac{1}{\bar{\tau}(x)} = 10^5\, x^2 \qquad (3.9)$$

which means that a phonon with $x = 1$ has a mean free path of the order of centimeters (i.e. dimensions of the sample). Figures III.1 and III.2 show plots of v_1 and δ_1 versus ω respectively for different values of

the applied A.C. field. We see from the figure that much below the frequency $\omega_c \sim 10^8$ sec^{-1} the amplitude of the drift velocity is nearly independent of ω while much above this frequency the drift current decreases to the value $\mu_{p.e.}F$. $\mu_{p.e.}$ 25000 cm/volt.sec. evaluated numerically from eq. (1.18a). The phase lag between the applied field and the current has a maximum in the vicinity of ω_c.
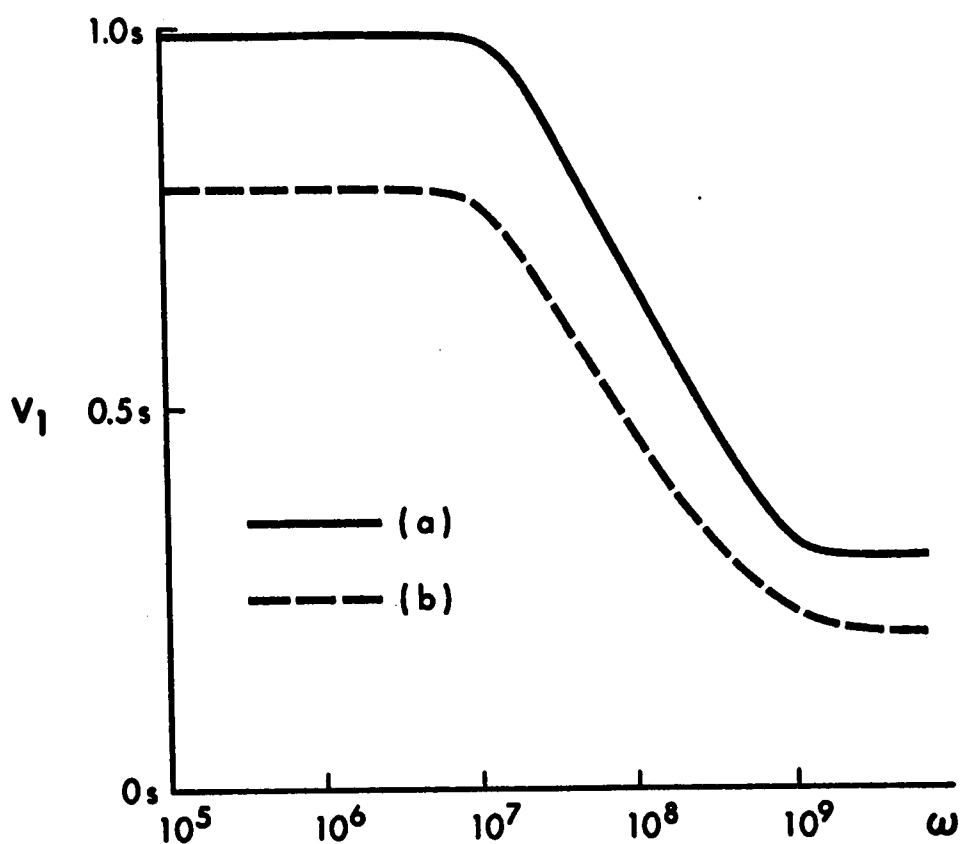
Figure III.1 displays v_1 versus ω for two values of the amplitude of the applied field, (a) 12 volts and (b) 8 volts.
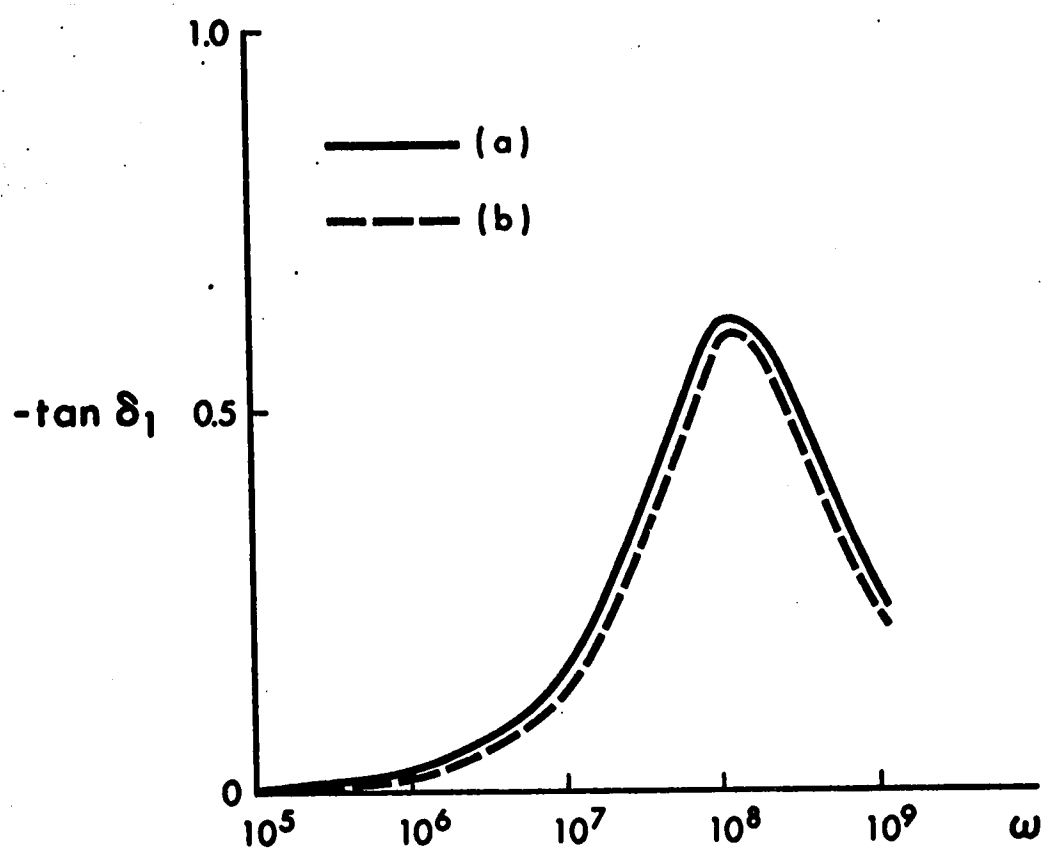
Figure III.2 displays δ_1 versus ω for the same applied fields.

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