Solid-State Electronics, 1978, Vol. 21, pp. 259–266. Pergamon Press. Printed in Great Britain

HIGH-FIELD ELECTRONIC CONDUCTION IN INSULATORS

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Abstract—The quantum theory of electronic transport phenomena in large electric fields in highly dissipative media is critically examined. Serious conceptual problems and computational difficulties arise because neither the field nor the dissipation can be treated as a perturbation. We review a decade-old calculation of the velocity acquired by an electron in a finite electric field in a polar crystal and subsequent work which expanded our understanding of our method and results. A key feature of the earlier work was that in a single curve of electric field vs velocity, all the expected phenomena appeared, including a threshold field for producing hot electrons, in quantitative agreement with experiment, and a decreasing rate of energy loss with velocity for very fast electrons. A more recently studied problem, that of electron acceleration below the threshold field will be discussed. This problem is very important since such acceleration is the necessary precursor of ionization and breakdown. The physical significance of dissipation processes far from thermal equilibrium will also be mentioned.

1. INTRODUCTION

The purpose of this review is to discuss several general calculations of electronic transport in insulators at high electric fields[1–7]. While we are motivated by problems arising in the study of device physics[8, 9], these problems are discussed here in the general framework of insulator physics. Focusing on the primary differences between insulators and semiconductors, we examine the necessity of using somewhat more involved analytic methods in order to approach these problems without making unsupportable approximations. We then consider the problem of the steady-state velocity acquired by an electron in a finite (subthreshold) electric field in a dissipative medium, e.g. polar insulator. Here the effectiveness of the material to dissipate the energy an electron acquires in falling through a large electric field leads to a very interesting steady-state problem requiring a fully quantum mechanical treatment. While high electric fields and dissipative media are usually very difficult to treat in the Schroedinger or Heisenberg picture, they are readily included in the path-integral method developed by Feynman[10–15].

Having treated this steady-state problem, we turn to another subthreshold problem, that of the transition rate of electrons from quasi-steady states to accelerating states. This "tunneling in momentum space" can provide a source of hot electrons for ionization (and hence breakdown) or for cold-cathode emission. A simple expression is given to determine the relative acceleration efficiencies of various materials. For reasons of simplicity the acceleration problem is discussed for weak coupling in the context of the Boltzman equation. A discussion for more realistic coupling can be found elsewhere[7]. We also note several problems associated with the relaxation approximation and scattering rates in applied fields.

Topics discussed are, of course, only a very small sample of the multitude of interesting problems presented by dynamic, steady-state phenomena well away from thermal equilibrium[16, 17]. As these problems become amenable to solution, we shall be in a much better position to understand the role of intrinsic insulator properties on the fundamental limitations of electronic devices.

2. GENERAL FEATURES

The semiconductor physicist tends to picture an insulator as a semiconductor with a large (>3 eV) band gap, somewhat hard to grow in a crystalline, nearly impurity- and defect-free form. While such a picture may be convenient at times, it is often more appropriate to regard the insulator, as a distinct entity, that is, as one of the three states of crystalline solids. While this is certainly the case for electronic properties such as surface states, photo emission, and optical absorption[18], it is also the case for electronic conduction, as we shall see.

Consider first the concept of a Fermi level, which, together with the concepts of a band gap and of electrons and holes, is one of the most important conceptual tools in semiconductor physics. The Fermi level can play only a minor role in discussing insulators for the following reason. If the energy level of the trapping state is more than about 50 kT (1.25 eV at room temperature) away from a band edge, then the dependence of the Fermi level E_F on the trapping level E_L becomes inordinately large even for otherwise negligibly small concentrations. For example, consider a level above the Fermi level: $E_V < E_F < E_L < E_C$. Assuming for simplicity that $N_V = N_C = N$, a simple calculation yields

$$\gamma \equiv \partial E_F/\partial E_L = \frac{1}{2}(1 + (N/N_L)\exp -(E_C - E_L)/kT)^{-1}. \tag{2.1}$$

Ordinarily in semiconductors $\gamma \ll 1$ at room temperature. However, suppose $E_C - E_L = 1.4$ eV and $N/N_L = 10^{23}/\text{cm}^3$ (1 impurity per cm^3), then $\gamma = 0.48$, barely less than the maximum value of 0.5. Now it is clear that one impurity or defect per cubic centimeter cannot affect physical properties at room temperature in

any significant manner. It follows that any entity which is strongly dependent on such a variable, cannot enter into the physics of the problem. Mention need hardly be given to what the above implies about the utility of an E_F in the presence of "natural" defects randomly spread throughout the material.

Whether or not the problem of obtaining high-purity insulators is intrinsic, owing to the large band gap and ionicity usually present, or merely fabricational, if one postulates a pure insulator other differences from semiconductors immediately arise. The most important of these is electron–phonon scattering, especially optical phonons in insulators with large ionic polarizabilities[19]. The coupling constant α rises from 0.02–0.04 in such "ionic" semiconductors as InSb and GaAs to 1–3 in SiO_2 and Al_2O_3. Also in these large gap materials the band-effective mass is of the order of the free electron mass. Mean free paths then fall to the order of Angstrom or tens of Angstroms. This stronger scattering in turn implies higher fields are needed to accelerate the electrons. The moderately large scattering renders perturbation theory and quasi-particle approaches unworkable, while the Boltzman equation can be expected to give a qualitative picture at best. In addition we note in Appendix A how the presence of the electric field alters the scattering rate usually obtained from Fermi's Golden Rule.

Concerning the Boltzman equation, while it is well-known that in its usual form

$$\frac{\partial f(\mathbf{p}, t)}{\partial t} + \mathbf{F}_t \cdot \frac{\partial f(\mathbf{p}, t)}{\partial \mathbf{p}} = \sum_{\mathbf{p}'} [R(\mathbf{p}, \mathbf{p}')f(\mathbf{p}', t) - R(\mathbf{p}', \mathbf{p})f(\mathbf{p}, t)] \tag{2.2}$$

the scattering rate $R(\mathbf{p}, \mathbf{p}')$ from $\mathbf{p}'$ to $\mathbf{p}$ must be weak[20], $\alpha \ll 1$, what is not widely appreciated is that the electric force $\mathbf{F}_t$, must also be weak[7] and very slowly time varying[21]. Provided the scattering remains weak, oscillatory $\mathbf{F}_t$ necessitate a convolution of a scattering function and $f(\mathbf{p}, t)$[7, 21], while a large, static $\mathbf{F}$, as mentioned above, introduces, among other things, a modification in R[7]. Finally, while a relaxation-time approximation is often made for the right-hand side of (2.2), for optical phonon scattering it is neither accurate nor necessary (Appendix B) and leaves out important physics (Section 4).

From this brief summary it is clear that the study of electronic transport, especially in the presence of high fields, requires a somewhat different point of view than a similar study in semiconductors. This will be apparent in the theoretical work reviewed here. The point to be made at this juncture is that the additional complexities which enter when treating insulators necessitate the more involved treatment which follows.

3. HIGH-FIELD TRANSPORT, DRIFT

My initial interest in the general problem of the motion of an electron in a dissipative medium under the influence of finite electric and magnetic fields arose in attempting to understand theoretically the very high rate of loss of energy (0.03 eV/Å) observed in Al–Al_2O_3–Al, cold-cathode structures[8, 9]. While most scattering mechanisms could account for only a small fraction of this loss, it was possible that owing to the large ionic polarizability present ($\alpha = 3$) in the oxide, the electron could dissipate all the kinetic energy it acquired from the applied electric field to the longitudinal optic phonons, even for electric fields of the order of 3 MV/cm. A new type of calculation was necessary since the mean-free path of 4 Å and the mean-free time of 10^{-15} sec implied by such a high loss rate could not be handled realistically by existing perturbation methods.

Before embarking on any calculation, a model and a quantity to calculate must be chosen. In choosing a model we made a number of approximations. (1) We ignored the interaction of the electron with the static lattice potential. This is reasonable since scattering from a static lattice can result in no loss of energy, and since the electron's energy, even when heated, is expected to be less than about 0.2 eV. A lattice mass should, therefore, adequately encompass this effect. (2) We assumed that the electron couples only to the local polarization, which is first order in the ion's displacement. This is reasonable since at the energies of interest the electron's wave function is expected to be about 40 Å wide and, therefore, the fine details of the scattering potential are not seen. (3) We assumed that in the absence of the conduction electrons, the longitudinal optic phonons oscillate as perfect harmonic oscillators. This is reasonable since the IR absorption is reasonably sharp, at least when compared with the electron's mean-free time. (4) We ignored the interaction of the conduction electrons with each other as they are so few in number and as their mutual scattering can result in no total energy loss. (5) We also ignored the interaction of the conduction electrons with the electrons of the insulator. So long as the lattice is effective in dissipating the energy acquired from the field, the electron's energy will be at most 0.2 eV and, hence, incapable of ionizing the insulator's electrons. Thus this interaction is also non-dissipative and, therefore, of subsidiary importance. Questions of energetic electrons, ionization, etc., will be touched upon in Section 4. For the cold-cathode problem suffice it to say that the entire voltage drop across the device is insufficient to produce an electron capable of ionization. We should stress, however, that we made no approximation regarding the size of the coupling constant, temperature, or electric field.

Turning now to the quantity to be calculated, we approach the heart of the matter. The standard approach of calculating a distribution is unsatisfactory because the concept of a distribution is suspect. The electron is always entangled, so to speck, with the lattice: it never has a well-defined momentum as it does, for example, between collisions in weakly scattering material. We can determine the steady-state velocity as a function of the applied electric field by calculating the expectation value of the velocity operator in the presence of the field. For sufficiently large electric field, of course, no solution is possible, the lattice being no longer able to arrest the electron's acceleration. Such an approach was actually carried out originally[1], however, it involved an exces-

sive amount of intuition to arrive at a realistic result. This was primarily because the velocity operator $\hat{\mathbf{p}}/m$ contains no physics specific to the problem of interest the wave functions or density matrix used must, therefore, carry all the information about the solution. By contrast, by calculating the expectation value of the operator equation for the conservation of momentum

$$\dot{\hat{\mathbf{p}}} = i[H, \hat{\mathbf{p}}]/\hbar \tag{3.1}$$

one can obtain at once a relation for the field $\mathbf{E}$ in terms of the steady-state velocity $\mathbf{v}$, without demanding as specific a solution as needed in calculating $\langle\hat{\mathbf{p}}/m\rangle$. This is because in the commutator a gradiant of the potential specific to the problem arises, and, hence, less detail of the interaction is needed in the solution.

Knowing what to calculate, while essential, is of course not enough: one must know how to calculate as well. On this point we shall not go into great detail as adequately detailed treatments are available[2–7]. The essential features, however, must be outlined. The two principal difficulties in calculating the expectation value of (3.1) fully quantum mechanically are the presence of the finite electric field and the highly dissipative nature of the system. It is an outstanding feature of the Feynman path-integral approach[10–15] that both these important effects can be included routinely. Furthermore, the phonon coordinates can be eliminated exactly. While no approximation need be made regarding the electric field, the exact influence functional remaining after integrating out the phonons must be approximated; however, it can be approximated by a similar, dissipative, influence fluctional. Since the approximate solution (zero-order solution) already contains the basic features of the problem, the finite field and dissipation, we are again in a position to consider perturbation theory, this time in the difference between the exact and approximate influence functionals, both qualitatively similar to each other.

The result obtained for the expectation value of (3.1) under steady-state conditions is the following.

$$\mathbf{F} + \mathbf{v}\times\mathbf{H} = \sum_{n,\mathbf{k}} \mathbf{k}|C_{\mathbf{k},n}|^2 \int_{-\infty}^{\infty} d\xi T_{\omega_{\mathbf{k},n}}(\xi)\, e^{-i\mathbf{k}\cdot\mathbf{v}\xi} e^{-\mathbf{k}\cdot\bar{\mathbf{L}}(\xi)\cdot\mathbf{k}} \tag{3.2}$$

where $\mathbf{F} = q\mathbf{E}$ the electric force, $\mathbf{H}$ is the magnetic field, and

$$T_\omega(\xi) = e^{i\omega\xi}(1 - e^{-\beta\omega})^{-1} + e^{-i\omega\xi}(e^{\beta\omega} - 1)^{-1} \tag{3.3a}$$

$$\bar{\mathbf{L}}(\xi) = \int_{-\infty}^{\infty} \frac{d\nu}{2\pi i} \frac{1}{\mathbf{Z}_\nu} 4\pi i \mathbf{G}(-\nu) \frac{1}{\mathbf{Z}_\nu^\dagger} (1 - e^{i\nu\xi}) \tag{3.3b}$$

$$\mathbf{Z}_\nu = -\mathbf{m}(\nu + i\epsilon)^2 - i(\nu + i\epsilon)\underline{\epsilon}\mathbf{H} + 4\int_0^\infty d\xi (1 - e^{i\nu\xi}) Im(\mathbf{G}^*(\xi)) \tag{3.3c}$$

$$\mathbf{G}^*(\xi) = \frac{1}{2}\sum_{n,\mathbf{k}} |C_{\mathbf{k},n}|^2 \mathbf{k}\mathbf{k} T_{\omega_{\mathbf{k}}}(\xi)\, e^{-i\mathbf{k}\cdot\mathbf{v}\xi} e^{-\mathbf{k}\cdot\bar{\mathbf{L}}(\xi)\cdot\mathbf{k}} \tag{3.3d}$$

$$\mathbf{G}(\nu) = \int \frac{d\xi}{2\pi} e^{i\nu\xi} \mathbf{G}(\xi) \tag{3.3e}$$

all of which arise from the Hamiltonian

$$H = \frac{1}{2}(\mathbf{p} - q\mathbf{A}/c)\mathbf{m}^{-1}(\mathbf{p} - q\mathbf{A}/c) - q\mathbf{E}\cdot\mathbf{x} - q\mathbf{e}_t\cdot\mathbf{x} + \sum_{n,\mathbf{k}} \hbar\omega_{\mathbf{k},n} a^\dagger_{\mathbf{k},n} a_{\mathbf{k},n} + V^{-1/2}\sum_{n,\mathbf{k}} (C_{\mathbf{k},n} a_{\mathbf{k},n}\, e^{i\mathbf{k}\cdot\mathbf{x}} + C^*_{\mathbf{k},n} a^\dagger_{\mathbf{k},n}\, e^{-i\mathbf{k}\cdot\mathbf{x}}) \tag{3.4}$$

setting $\hbar = c = V = 1$, $\mathbf{m}$ is the fixed lattice mass mentioned above; $\beta = 1/kT$, T being the lattice temperature. Here $\omega_{\mathbf{k},n}$ is the dispersion of the nth phonon branch at wave vector $\mathbf{k}$, $\mathbf{A}$ is the vector potential corresponding to the constant magnetic field $\mathbf{H}$, V is the volume of the system. Expressions (3.3b–e) are a self-consistent set of equations from which $\bar{\mathbf{L}}(\xi)$ can be obtained by iteration, for example.

Certain physical aspects of the derivation of (3.2) should be mentioned. Prior to introducing the approximate influence functional, the electronic system is shifted to a coordinate system translating with velocity $\mathbf{v}$, the steady-state velocity of the electron: $\mathbf{x}_t = \mathbf{y}_t + \mathbf{v}t$. In this drifting system the above mentioned approximation is made; that is, the exact influence functional is replaced by a distribution of oscillators characterized by $\mathbf{G}(\nu)$, the oscillator strength at frequency ν. In order to determine this strength, which is a function of $\mathbf{v}$ and $\mathbf{H}$ and hence $q\mathbf{E} = \mathbf{F}$, a small probe field $\mathbf{e}_t$ is applied in the drifting frame. From the a.c. response to this probe field the dynamic self-consistency of (3.3b–e) for $\bar{\mathbf{L}}$, $\mathbf{Z}$ and $\mathbf{G}$ arises. Requiring this self-consistency yields approximate solutions which satisfy several optical sum rules[22, 23] as well as more stringent frequency-by-frequency criteria[5, 6]. Such solutions also simplify higher order perturbation expressions[7]. The above mentioned connection between the small-signal a.c. response in the drifting frame and the self-consistent influence functional representing the dissipative system for the drifting electron in a finite $\mathbf{F}$ represents yet another novel feature of this approach.

Returning to the result (3.2) itself, for $\mathbf{H} = 0$ we obtain a relation between the electric force $\mathbf{F}$ on the electron and its expectation steady-state velocity. In this single expression is contained all the physical features expected of the result including the nonlinearities inherent in this transport problem. We obtained with increasing velocity first the strongly temperature dependent, low-field mobility, as found by FHIP[24] in the limit of zero frequency of the applied field. Then, for initial lattice temperature below the reststrahlen energy, there is a rapid increase in the rate of energy loss as the electron's translational kinetic energy approaches this optical-phonon energy. This is followed by a temperature-independent threshold $\mathbf{F}_T$, or maximum loss of energy with distance. For $\mathbf{F} > \mathbf{F}_T$ no steady-state velocity is possible. Finally, as the velocity increases further, a temperature-independent decrease in energy loss as $\ln(\nu)/\nu^2$ is obtained as expected. For initial lattice temperatures above the optical phonon energy, the low-field linear region passes smoothly through a broad temperature-

dependent threshold which portrays the dominance of scattering from existing optical phonons. For very large velocities, the weak coupling limit of the solution gives the expected perturbation result. Thus, in a single family of field-velocity curves with temperature as the parameter, all the expected physical phenomena appear. Agreement with experiment is obtained for phenomena occuring in the vicinity of the threshold. We concluded that optical-phonon scattering can produce the large loss of electron energy observed in tunnel-emission devices.

What is of greater interest in the cold-cathode problem is the number of electrons accelerated to high energies, several eV. Such a calculation done on the level of the d.c. and a.c. transport problems discussed above is beyond the scope of this review. Fortunately a simple derivation, qualitatively accurate, can be given. This we present in Section 4.

4. HIGH-FIELD TRANSPORT, ACCELERATION

While I shall not develop the point, any article on high-field conduction in insulators is expected to say something about electronic breakdown. Such breakdown is often pictured as resulting from a (slow or fast) sustained ionization or even avalanche multiplication. The only comment I wish to add to this picture is to suggest that, since on the one hand accelerating electrons must overcome an energy-loss barrier at about 0.1 eV and on the other hand must accelerate to 10 eV before impact ionization can occur, the problem should be treated in two parts: (1) the acceleration to ionizing energies, and (2) the ionization process itself. In nearly all cases (2) is treated in detail while hardly any attention is given to (1). In this section (1) will be considered in some detail.

It should come as no surprise that when an electric field in excess of the threshold field is applied to an insulator, any electron in the conduction band of the insulator will accelerate to ionization energies, the lattice being unable to dissipate the energy acquired from the field. What I have not seen stated before is that for applied, *sub*threshold fields there is still a probability that an electron can accelerate to ionizing energies. Such a possibility can come about as follows. If the electron can go without scattering for a sufficiently long time that it can accelerate to a velocity at which the scattering rate has passed its maximum and greatly diminished, then it can continue to accelerate with only a small probability of further energy loss. For small applied fields, the chance for this to occur is, of course, very small. As the field approaches the threshold field, however, the probability approaches unity. Thus, whereas classically the probability goes abruptly from 0 to 1 in passing through the threshold field, quantum-mechanically a continuous transition is possible. Because of its vague analogy to tunneling through an energy barrier in position space, we can refer to this phenomena as a tunneling in momentum space. Unfortunately, no analogy in calculation carries over.

For the purposes of this article it will suffice to treat this phenomena using the Boltzman equation (2.2). A treatment on the level of Section 3 is extremely involved and at present not nearly as well-developed[7]. The Boltzman approach will, of course, apply directly to weak-scattering problems as commonly arise in semiconductors. The real reason, however, is to exhibit this new phenomenon as clearly as possible without sacrificing the important qualitative features of the problem. The general idea is this. We shall calculate the rate at which electrons transition from low energy, quasi-steady states to accelerating states of relative high energy by calculating the time rate of change of the electron distribution function at high energy. The latter, in turn, will be expressed in terms of the quasi-static distribution at low energy, which can be calculated by standard means.

Thus, we begin by writing (2.2) for a constant field.

$$\frac{\partial f(\mathbf{p},t)}{\partial t}+\mathbf{F}\cdot\frac{\partial f(\mathbf{p},t)}{\partial \mathbf{p}}=\sum_{\mathbf{p}'}[R(\mathbf{p},\mathbf{p}')f(\mathbf{p}',t)-R(\mathbf{p}',\mathbf{p})f(\mathbf{p},t)]. \tag{4.1}$$

Ordinarily one sets $f(\mathbf{p},t)=f(\mathbf{p})$ and ignores the first term. As we shall see, this is *not* correct except under unusual circumstances. The driving force $\mathbf{F}$, however small and even though static, eventually drives the distribution to higher energies. This is readily seen if we solve (4.1) by the method of characteristics[25]. If we define

$$1/\tau_{(\mathbf{p})}\equiv\sum_{\mathbf{p}'}R(\mathbf{p}',\mathbf{p}) \tag{4.2}$$

and consider the variables; $t(s)$, $\mathbf{p}(s)$;

$$\mathrm{d}/\mathrm{d}s=\mathrm{d}t/\mathrm{d}s\,\partial/\partial t+\mathrm{d}\mathbf{p}/\mathrm{d}s\,\partial/\partial\mathbf{p} \tag{4.3a}$$

so that

$$\mathrm{d}t/\mathrm{d}s=1,\quad t(s)=(s-s_0)+t_0 \tag{4.3b}$$

$$\mathrm{d}\mathbf{p}/\mathrm{d}s=\mathbf{F},\quad \mathbf{p}(s)=\mathbf{F}(s-s_0)+\mathbf{p}_0, \tag{4.3c}$$

then (4.1) becomes

$$\frac{\mathrm{d}f(\mathbf{p}(s),t(s))}{\mathrm{d}s}+\frac{f(\mathbf{p}(s),t(s))}{\tau(\mathbf{p}(s))}=\sum_{\mathbf{p}'}R(\mathbf{p}(s),\mathbf{p}')f(\mathbf{p}',t(s)). \tag{4.4}$$

Solving (4.4) as an ordinary differential equation in s, we find

$$f(\mathbf{p}(s),t(s))=f(\mathbf{p}(s_0),t(s_0))\exp\left[-\int_{s_0}^{s}\mathrm{d}s''/\tau(\mathbf{p}(s''))\right]$$
$$+\int_{s_0}^{s}\mathrm{d}s'\exp\left[-\int_{s'}^{s}\mathrm{d}s''/\tau(s'')\right]\sum_{\mathbf{p}''}R(\mathbf{p}(s'),\mathbf{p}'')f(\mathbf{p}'',t(s')). \tag{4.5}$$

Relations (4.3b,c) specify a collection of characteristics in $(\mathbf{p},t)$. By choosing the appropriate characteristic, we can recover $f(\mathbf{p},t)$ from $f(\mathbf{p}(s),t(s))$. Choosing $s_0=t_0$ yields $t(s)=s$. Relabelling s by t converts (4.5) into

$$f(\mathbf{p},t)=f(\mathbf{p}-\mathbf{F}(t-t_0),t_0)\exp\left[-\int_{t_0}^{t}\mathrm{d}t''/\tau(\mathbf{p}-\mathbf{F}(t-t''))\right]$$
$$+\int_{t_0}^{t}\mathrm{d}t'\exp\left[-\int_{t'}^{t}\mathrm{d}t''/\tau(\mathbf{p}-\mathbf{F}(t-t'))\right]$$
$$\times\sum_{\mathbf{p}''}R(\mathbf{p}-\mathbf{F}(t-t'),\mathbf{p}'')f(\mathbf{p}'',t') \tag{4.6}$$

which describes the time evolution of $f(\mathbf{p}, t)$ under the influence of the applied field $\mathbf{F}$.

Before proceeding we must understand the physics expressed in (4.6). The first term is the transient term. It states that time t an electron initially at $\mathbf{p}-\mathbf{F}(t-t_0)$ has arrived at $\mathbf{p}$ without scattering, an event which occurs with transition probability $P(\mathbf{p}; t, t_0)$, where

$$P(\mathbf{p}; t, t') \equiv \exp\left[-\int_{t'}^{t} dt''/\tau(\mathbf{p}-\mathbf{F}(t-t''))\right]. \quad (4.7a)$$

While the transient term generally plays only a negligible role in the acceleration problem, we note that a similar transition probability enters into the second term in (4.6). This term states that if an electron at $\mathbf{p}'$ scatters at t' into $\mathbf{p}-\mathbf{F}(t-t')$, then at t it will arrive at $\mathbf{p}$ with probability $P(\mathbf{p}; t, t')$. It should be noted that had (4.1) been solved at the relaxation-time approximation only the relatively unimportant transcient term would arise.

If we take $\mathbf{F} = F\hat{\mathbf{x}}$, then we can write

$$P(\mathbf{p}; t, t') \equiv \exp\left[-\int_{p_x-F(t-t')}^{p_x} dp_x''/F\tau(p_x'', \mathbf{p}_\perp)\right]. \quad (4.7b)$$

Written this way it is evident that the probability P that an electron injected at rest can transition from a quasi-steady state to acceleration state is

$$P_0 = \exp\left[-\int_0^\infty dp_x''/F\tau(p_x'', 0)\right]. \quad (4.8)$$

This simple result has the following significance. If the integral in the exponent diverges, then the scattering rate is sufficient to effectively prohibit transitions to accelerating states. In this case one is justified in dropping the time derivative in (4.1), and a steady-state solution is valid. Naturally a cold-cathode could not be made from material which renders $P_0 = 0$. If the integral in (4.8) is finite, then transitions are possible. Knowing the maximum F at which a specific insulator can be operated and the scattering rate $1/\tau(\mathbf{p})$, one can calculate P_0 for each insulator of interest in order to determine the most efficient accelerator of electrons.

Returning now to (4.6), an integral equation for $f(\mathbf{p}, t)$, we must now determine the actual transition rate to accelerating states. Let $F^{>}(p_x', t)$ be the probability at t that $p_x > p_x'$. Take $t_0 = 0$. Clearly

$$F^{>}(p_x', t) = \int_{p_x'}^{\infty} dp_x \int d\mathbf{p}_\perp f(\mathbf{p}, t). \quad (4.9a)$$

A more convenient form for $f(\mathbf{p}, t)$ can be obtained by using (4.7b) in place of (4.7a) in (4.6) and by changing the p_x variable in (4.9a) to $p_x' = p_x - Ft$ when integrating the transcient term and to $p_x' = p_x - F(t-t')$ for the second term. It follows that

$$F^{>}_{(p_x', t)} = \int_{p_x'-Ft}^{\infty} dp_x \int d\mathbf{p}_\perp f(\mathbf{p}, 0) P(p_x + Ft, p_x; \mathbf{p}_\perp) + \int_0^t dt' \int_{p_x'-F(t-t')}^{\infty} dp_x \int d\mathbf{p}_\perp \sum_{\mathbf{p}'} R(\mathbf{p}, \mathbf{p}') f(\mathbf{p}', t') \times P(p_x + F(t-t'), p_x; \mathbf{p}_\perp) \quad (4.9b)$$

$$P(p_2, p_1; \mathbf{p}_\perp) \equiv \exp\left[-\int_{p_1}^{p_2} dp_x/F\tau(p_x, \mathbf{p}_\perp)\right]. \quad (4.9c)$$

Where we have in addition replaced p_x' by p_x. For reasons that will become evident shortly, we must further manipulate the $\int dt' \int dp_x$ in (4.9b). Thus

$$\int_0^\infty dt' \int_{p_x'-F(t-t')}^{\infty} dp_x$$

$$= \int_0^t dt' \int_{p_x'-Ft}^{\infty} dp_x u[p_x - (p_x' - F(t-t'))]$$

$$= \left[\int_{p_x'}^{\infty} dp_x + \int_{p_x'-Ft}^{p_x'} dp_x\right] \times \int_0^t dt' u[p_x - (p_x' - F(t-t'))].$$

If we choose p_x' sufficiently large that $R(\mathbf{p}, \mathbf{p}')$ is negligible, then we can ignore the p_x' to ∞ integration in p_x. If we let $\tau = t' + (p_x' - p_x)/F$, then changing from dt' to $d\tau$ above we find

$$= \int_{p_x'-Ft}^{p_x'} dp_x \int_{(p_x'-p_x)/F}^{t} d\tau$$

$$= \int_{p_x'-Ft}^{p_x'} dp_x \int_0^t d\tau u[\tau - (p_x' - p_x)/F]$$

$$= \int_0^t d\tau \int_{p_x'-F\tau}^{p_x'} dp_x.$$

We may now write $F^{>}$ in the form

$$F^{>}(p_x', t) = \int_{p_x'-Ft}^{\infty} dp_x \int d\mathbf{p}_\perp f(\mathbf{p}, 0) P(p_x + Ft, p_x; \mathbf{p}_\perp) + \int_0^t d\tau \int_{p_x'-F\tau}^{p_x'} dp_x \int d\mathbf{p}_\perp \sum_{\mathbf{p}'} R(\mathbf{p}, \mathbf{p}') \times f(\mathbf{p}', \tau - (p_x' - p_x)/F) P(p_x' + F(t-\tau), p_x; \mathbf{p}_\perp) \quad (4.9d)$$

from which the transition rates of interest can be determined.

Returning to the problem in physical terms, what we have is a distribution of electrons which for the most part remain in a quasi-steady state, translating through the insulator with some average velocity $\mathbf{v}$, as calculated in Section 3. Leaking off from this distribution into accelerating states is a certain fraction $F^{>}$. If we call the quasi-steady state distribution $f^{qss}(\mathbf{p}, t)$, then the rate per electron at which electrons transition from the quasi-steady state to the accelerating state is simply

$$r(F) \equiv dF^{>}(p_x', t)/dt \Big/ \sum_{\mathbf{p}} f^{qss}(\mathbf{p}, t) \equiv 1/\tau_d(F). \quad (4.10)$$

After initial transcients, we assume that $f(\mathbf{p}, t)$ for the electrons in the quasi-steady state $f^{qss}(\mathbf{p}, t)$ has the form $f^{qss}(\mathbf{p}) \exp(-t/\tau_d)$. Then using (4.6) $f^{qss}(\mathbf{p})$ satisfies

$$f^{qss}(\mathbf{p}) = \int_{-\infty}^{p_x} \frac{dp_x'}{F} \exp\left[-\int_{p_x'}^{p_x} \frac{dp_x''}{F}\left(\frac{1}{\tau(p_x'', \mathbf{p}_\perp)} - \frac{1}{\tau_d}\right)\right] \times \sum_{\mathbf{p}''} R(p_x', \mathbf{p}_\perp, \mathbf{p}'') f^{qss}(\mathbf{p}'') \quad (4.11)$$

and we obtain for $r(F)$

$$r(F) = \sum_{\mathbf{p}} f^{qss}(\mathbf{p})/\tau_d(p) \Big/ \sum_{\mathbf{p}} f^{qss}(\mathbf{p}) = 1/\tau_d(F) \tag{4.12a}$$

$$1/\tau_d(p) = \int_{-\infty}^{\infty} d\mathbf{p}' R(\mathbf{p}', \mathbf{p}) \exp[(p_x' - p_x')/F\tau_d)] P(\infty, p_x'; \mathbf{p}_\perp). \tag{4.12b}$$

Here $1/\tau_d(\mathbf{p})$ has the meaning of the transition rate of an electron in state $\mathbf{p}$. Having $F^>$ in the form of (4.9d), the time derivative needed for $r(F)$ is further simplified by noting that

$$P(p_x' + F(t-\tau), p_x; \mathbf{p}_\perp) \approx P(p_x', p_x; \mathbf{p}_\perp)$$

for $0 \leq \tau \leq t$ by the way in which p_x' was chosen. Thus the very physical result (4.12a) emerges. (The reader not appreciating this point is invited to evaluate (4.10) using alternative expressions for $F^>$.)

For small fields F, τ_d can be expected to be rather large so that $\tau_d(\mathbf{p})$ is relatively independent of τ_d, and hence $\tau_d(p)$ can be calculated with*out* knowledge of the distribution. (Note that (4.12b) does not contain $f(\mathbf{p}, t)$ explicitly.) For larger fields τ_d will become appreciable, hence one must solve for τ_d self-consistently between (4.11) and (4.12a,b). The simplest procedure is to assume $\tau_d = \infty$ in (4.11) and (4.12b), get a τ_d from (4.12a), etc. To be sure, this expression is most meaningful when $F\tau_d \gg p_x'$, which is usually the case somewhat below threshold. Excellent solutions of (4.9) exist for $\tau_d = \infty$[26], and their modification for $\tau_d < \infty$ should be straightforward; for example, replace $\partial f(\mathbf{p}, t)/\partial t = 0$ by $\partial f(\mathbf{p}, t)/\partial t = -f(\mathbf{p}, t)/\tau_d$.

The foregoing, while providing a clear physical picture of the problem of electron acceleration below threshold, is nonetheless based on the Boltzman equation and hence is valid only for small electron-lattice coupling and small electric fields. Elsewhere[7] I have generalized the above derivation to include the modification of propagation and scattering due to larger electric fields and electron interaction with the solid. However these modifications are only expected to be useful for coupling constants $\alpha < 1$. For $\alpha > 1$ a direct evaluation of $1/\tau_d(\mathbf{p})$ may be more meaningful[7].

5. CONCLUSION

In this brief review we have touched on several general features of insulators which distinguish them from semiconductors, outlined the derivation of the relation between the drift velocity and electric field at high electric fields where the relation is nonlinear, and introduced the problem of electron transitions from low-energy, quasi-steady states to high-energy accelerating states at high but subthreshold fields. The latter was regarded as a necessary precursor to ionization and other electronic breakdown phenomena. Looking toward the future we must keep in mind that significant energy loss, dissipation, is intrinsic in insulators, necessitating the high fields applied. Calculations of transport properties, therefore, must of necessity be such as to be valid under such conditions. By contrast, standard approaches to transport are usually geared to problems in which dissipation is a small perturbation. For insulators this is not realistic, and methods, possibly as outlined above, will have to become more widely used in order to understand the more sophisticated experiments currently underway[27, 28]. Finally we note that the nature of systems under impressed fields, that is with energy flowing in from a field and out through dissipation, is potentially much more interesting to study than that of the same systems at or near thermal equilibrium[16, 17].

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APPENDIX A

Scattering rates in an electric field

In this appendix we calculate the scattering rate for electron-phonon scattering in the presence of an arbitrary, constant electric field $\mathbf{E}$. In lowest order (zero order) the electron is assumed to be a free particle accelerating in the applied field:

$$H_0 = \mathbf{p}^2/2m - \mathbf{F} \cdot \mathbf{x}, \quad \mathbf{F} = q\mathbf{E}. \tag{A1}$$

We shall examine the lowest order of scattering (first order) between the electron and the phonon bath in which it moves. The total Hamiltonian is as given in (3.4).

As mentioned in the text, the Feynman path-integral method can handle H_0 exactly and conveniently, especially when compared to the Airy-function solution which would arise in a Schrodinger approach. If we use the result of a previous calculation[3] for the scattering rate, and specialize this result to the free-particle H_0 of (A1) we obtain for $R(\mathbf{p}', \mathbf{p})$, the rate at which electrons are scattered from $\mathbf{p}$ to $\mathbf{p}' = \mathbf{p} - \hbar\mathbf{k}$

$$R(\mathbf{p}', \mathbf{p}) = |C_{\mathbf{k}}|^2 2\mathrm{Re}\left[\int_0^\infty dt \left(\frac{e^{i\omega_{\mathbf{k}} t}}{1 - e^{-\beta\hbar\omega_{\mathbf{k}}}} + \frac{e^{-i\omega_{\mathbf{k}} t}}{e^{\beta\hbar\omega_{\mathbf{k}}} - 1}\right) e^{i\hbar k^2 t/2m} e^{-i\mathbf{k}\cdot\mathbf{p}t/m} e^{i\mathbf{k}\cdot\mathbf{F}t^2/2m}\right]. \tag{A2}$$

IfF is zero, or otherwise negligible, we obtain

$$R(\mathbf{p}', \mathbf{p}) = \frac{2\pi}{\hbar} |C_{\mathbf{p}-\mathbf{p}'}|^2 [(1 - e^{-\beta\hbar\omega_{\mathbf{k}}})^{-1} \delta(\hbar\omega_{\mathbf{k}} - \hbar\mathbf{k}\cdot\mathbf{p}/m + \hbar^2 k^2/2m) + (e^{\beta\hbar\omega_{\mathbf{k}}} - 1)^{-1} \delta(-\hbar\omega_{\mathbf{k}} - \hbar\mathbf{k}\cdot\mathbf{p}/m + \hbar^2 k^2/2m)] \tag{A3}$$

the usual Fermi Golden Rule. For F non-negligible, the $\int d\tau$ which would normally yield an energy conserving delta function now yields a broadened and skewed function, which we shall examine briefly.

In (A2) the first term corresponds to the emission of a phonon of wave vector $\mathbf{k}$ and the second to an absorption of $-\mathbf{k}$. Consider the emission. The integral of interest J is

$$J = 2Re \int_0^\infty dt\, e^{iAt} e^{iBt^2/2} \tag{A4a}$$

$$A \equiv \hbar\omega_{\mathbf{k}} + \hbar^2 k^2/2m - \hbar\mathbf{k}\cdot\mathbf{p}/m = E_{\text{final}} - E_{\text{initial}} \tag{A4b}$$

$$B \equiv \hbar^2 \mathbf{k}\cdot\mathbf{F}/m. \tag{A4c}$$

We manipulate as follows

$$J = 2\left(\int_{-A/B}^\infty dt + \int_0^{-A/B} dt\right) \cos(At + Bt^2/2) \tag{A5a}$$

$$= 2\int_0^\infty dx \cos(Bx^2/2 - A^2/2B) - 2\int_0^{-A/B} dt \cos(At - Bt^2/2). \tag{A5b}$$

The first term in J integrates to

$$(\pi/B)^{1/2}[\cos(A^2/2B) + \sin(A^2/2B)] \to 2\pi\delta(A), \quad B \to 0$$

an even function of A. The second term can also be written

$$-2\int_0^{A/B} dx \cos(A^2/2B - Bx^2/2)$$

which is clearly odd in A and vanishes for $B = 0$.

While we shall not evaluate the second term in J (it can be expressed in terms of Fresnel integrals), we shall observe two important physical implications. (1) The broadening B implies an energy uncertainty ΔE in the scattering given by

$$\Delta E = B^{1/2} = \hbar(|\mathbf{k}\cdot\mathbf{F}|/m)^{1/2}. \tag{A6}$$

While ΔE is negligible for small F, it rises to about 0.1 eV in Al_2O_3 near threshold, 2.5 MV/cm, where $\mathbf{F} \| \mathbf{k}$. Note that no broadening arises if $\mathbf{F} \perp \mathbf{k}$. (2) The skewness introduced by the second term in J is also of interest. If $A/B > 0$, J is reduced; if $A/B < 0$, J is enhanced. For emission, where $B > 0$, $A/B > 0$ implies $E_{\text{final}} > E_{\text{initial}}$, while $A/B < 0$ implies $E_{\text{final}} < E_{\text{initial}}$. This is easily understood in physical terms. In emission, where the electron is scattered against the field, the field will absorb a portion of the electron's energy during the collision, and hence reduction in energy will be favored. The opposite effect, an enhancement in energy, can occur in absorption during those scattering events in which the electron scatters with the field. We conclude that the electric field can appreciably alter the scattering rates by broadening and skewing the energy conserving delta function in Fermi's Golden Rule.

APPENDIX B

Electron distribution in linear response

It is often expedient in solving the Boltzman equation (BE) to make use of the relaxation-time approximation to simplify the solution. However, even for small fields in which the change in the distribution is proportional to the field, considerable error in the nature of the distribution can be made if this approximation is involved. For low temperatures and optical phonon scattering, the typical argument runs as follows. We write $F = f_0 + f_1$, substitute into the BE, and obtain to first order in the field

$$\mathbf{F}\cdot\partial f_0(p)/\partial\mathbf{p} = \sum_{\mathbf{p}'} [R(\mathbf{p}, \mathbf{p}')f_1(\mathbf{p}') - R(\mathbf{p}', \mathbf{p})f_1(\mathbf{p})] \tag{B1}$$

where $f_0(p) = A\exp(-p^2/2mkT)$, $A = (2mkT\pi)^{3/2}$. Now we consider two regions in momentum space, $p < p_c$ and $p > p_c$ where $p_c = (2m\hbar\omega_L)^{1/2}$, where $\hbar\omega_L$ is the energy of an optical phonon. Now as we have seen already (A3) the scattering rate $R(\mathbf{p}, \mathbf{p}')$ contains both an emission and an absorption term so that either $p > p_c$ and $p' < p_c$ or $p < p_c$ and $p' > p_c$. Furthermore, emission is impossible if $p < p_c$. Thus in (B1) suppose $p < p_c$. Then the $R(\mathbf{p}, \mathbf{p}')f_1(\mathbf{p}')$ term involves scattering out to $p' > p_c$ by phonon absorption. If $f_1(\mathbf{p}')$ is sufficiently small the scattering in term can be ignored. If $p > p_c$, then the electron scattered in comes from $p' < p_c$. However, if by our previous argument for $p < p_c$

$$f_1(\mathbf{p}) = -\tau(p)\mathbf{F}\cdot\mathbf{p}f_0(p)/mkT, \tag{B2}$$

where

$$1/\tau(p) = \sum_{\mathbf{p}'} R(\mathbf{p}', \mathbf{p}), \tag{B3}$$

then

$$\sum_{\mathbf{p}'} R(\mathbf{p}, \mathbf{p}')f_1(\mathbf{p}') = 0, \quad p > p_c \tag{B4}$$

if $R(\mathbf{p}, \mathbf{p}') = R(\mathbf{p}, -\mathbf{p}')$, as it very nearly is. Thus for $p > p_c$ we also have (B2); in other words (B1) becomes

$$\mathbf{F}\cdot\partial f_0(p)/\partial\mathbf{p} = -f_1(\mathbf{p})/\tau(\mathbf{p}) \tag{B5}$$

the usual relaxation time approximation. As a check we return to $p < p_c$ to compare terms; the neglected term is again (B4) now with $p < p_c$. Thus the relaxation-time approximation seems consistent. Moreover note that as p passes from $p < p_c$ to $p > p_c$, $f_1(p)$ diminishes by the ratio of absorption rate to emission rate, which for $\exp(-\hbar\omega_L/kT) \ll 1$, the usual case, is very small. This means that $f_1(\mathbf{p})$ for $p > p_c$ is reduced from $f_1(\mathbf{p})$ for $p \approx 0^+$ not only by the Boltzman factor $\exp(-p^2/2mkT)$, but by $\tau(p_>)/\tau(p_<)$, where $p_>$ means $p > p_c$ and $p_<$ means $p < p_c$. Thus $f_1(\mathbf{p})$ is usually totally neglected for $p > p_c$. Physically this means that the source of $f_1(\mathbf{p})$ for all $\mathbf{p}$ is the electric field, scattering into the distribution being negligible.

The problem with the above argument is the relation $R(\mathbf{p}, \mathbf{p}') \approx R(\mathbf{p}, -\mathbf{p}')$, which while approximately true is not true to order $\exp(-\hbar\omega_L/kT)$. Thus physically while for $p_<$, the source of $f_1(\mathbf{p}<)$ is mainly the field, for $p_>$

$$\sum_{\mathbf{p}'} R(\mathbf{p}_>, \mathbf{p}')\tau(p')p'_z f_0(p') \gg p_{z>} f_0(p_>) \tag{B6}$$

($\mathbf{F} \| \hat{\mathbf{z}}$) and hence the source of $f_1(\mathbf{p}_>)$ is not the field at all but phonon absorption from $f_1(\mathbf{p}_<)$. Thus

$$f_1(\mathbf{p}_>) = \tau(p_>) \sum_{\mathbf{p}'} R(\mathbf{p}_>, \mathbf{p}')\tau(p')\mathbf{F}\cdot\mathbf{p}' f_0(p')/mkT \tag{B7a}$$

while

$$f_1(\mathbf{p}_<) = \tau(p_<)\mathbf{F}\cdot\mathbf{p}_< f_0(p_<)/mkT. \tag{B7b}$$

Now $f_1(p_>)/f_1(p_<) \approx f_0(p_>)/f_0(p_<)$ and $f_1(p_>)$ is not nearly so small as implied by the relaxation-time-approximation result. This difference does not affect the usual calculation of the mobility, but is essential in a conservation of momentum or conservation of energy approach.

It is of interest that (B1) can be solved exactly if one ignores $F_1(\mathbf{p})$ for $p > (4m\hbar\omega_L)^{1/2}$, which really is reasonable for $\exp(-\hbar\omega_L/kT) \ll 1$. The reason for this is that the energy exchange $|\Delta E| = \hbar\omega_L$. Hence exchanges of electrons are only possible between E and $E+\hbar\omega_L$, $E<\hbar\omega_L$, and E and $E-\hbar\omega_L$, $E>\hbar\omega_L$. Thus the problem divides into pairs of shells. Also because of the momentum dependence of the scattering, the angular distribution is sufficiently simple that $f_1(\mathbf{p})$ has the general form $g_1(p)\cos\theta_p$ for $\mathbf{F}\|\hat{\mathbf{z}}$. While we shall not carry out the calculation in detail, we shall focus on its main features.

(1) The rates given in (B1) are expressed in (A3). For optical-phonon scattering

$$|C_{\mathbf{p}-\mathbf{p}'}|^2 = \frac{\alpha B\hbar^3}{V|_{\mathbf{p}-\mathbf{p}'}|^2}, \quad B = \frac{4\pi\omega_L{}^2}{(2\pi)^3}\left[\frac{\hbar}{2m\omega_L}\right]^{1/2}. \tag{B8a}$$

$$|\mathbf{p}-\mathbf{p}'|^2 = p^2 + p'^2 - 2pp'\cos\gamma_{pp'}. \tag{B8b}$$

(2) In order to evaluate the sum over $\mathbf{p}'$ in the scattering-in term, it is convenient to pass to a coordinate system in which the $\hat{\mathbf{z}}$ axis is taken along $\mathbf{p}$. Then $\gamma_{pp'}$ in (B8b) becomes $\theta_{p'}$, while expanding $f_1(\mathbf{p})$ in the frame where $\mathbf{F}\|\hat{\mathbf{z}}$,

$$f_1(\mathbf{p}) = \sum_{l=0}^{\infty} g_l(p)P_l(\cos\gamma_p), \tag{B9a}$$

and then for $f_1(\mathbf{p}')$ passing to the $\hat{\mathbf{z}}\|\mathbf{p}$ frame, we have

$$P_l(\cos\gamma_p') = P_l(\cos\theta_p)P_l(\cos\theta_p') + 2\sum_{m=1}^{l}\frac{(l-m)!}{(l+m)!} \times P_l^m(\cos\theta_p)P_l^m(\cos\theta_p')\cos m(\phi_p - \phi_p'). \tag{B9b}$$

In doing the $\Sigma_{\mathbf{p}'}$, the $\cos m(\phi_p - \phi_p')$ will integrate to zero leaving only the first product. Since this decouples (B1) into equations for $g_l(p)$, one for each l, and since only $l=1$ is driven by $\mathbf{F}$,

$$f_1(\mathbf{p}) = g_1(p)\cos\theta_p. \tag{B9c}$$

(3) Carrying out the integrals over $\theta_{p'}$, $\phi_{p'}$ and using the delta functions to integrate $d_{p'}$, one obtains

$$\frac{Fp_< f_0(p_<)}{mkT} = g_1(p_>)\frac{2\pi B\alpha m p_>/(p_<{}^2+p_>{}^2)}{1-\exp(-\hbar\omega_L/kT)}J(s) - g_1(p_<)\frac{2\pi B\alpha m p_>/(p_<{}^2+p_>{}^2)}{\exp(\hbar\omega_L/kT)-1}I(s) \tag{B10a}$$

$$\frac{-Fp_> f_0(p_>)}{mkT} = g_1(p_<)\frac{2\pi B\alpha m p_</(p_<{}^2+p_>{}^2)}{\exp(\hbar\omega_L/kT)-1}J(s) - g_1(p_>)\left[\frac{2\pi B\alpha m p_</(p_<{}^2+p_>{}^2)}{1-\exp(-\hbar\omega_L/kT)} + \frac{2\pi B\alpha m p_>/(p_>{}^2+p_<{}^2)}{\exp(\hbar\omega_L/kT)-1}\right]I(s) \tag{B10b}$$

$$s \equiv 2p_<p_>/(p_<{}^2+p_>{}^2) \tag{B10c}$$

$$I(s) \equiv \int_{-1}^{1} d\mu(1-s\mu)^{-1} = \ln\left(\frac{1+s}{1-s}\right)\Big/ s \tag{B10d}$$

$$J(s) \equiv \int_{-1}^{1} d\mu\mu(1-s\mu)^{-1} = \left[\ln\left(\frac{1+s}{1-s}\right) - 2s\right]\Big/ s^2 \tag{B10e}$$

$$p_> = (p_<{}^2 + 2m\hbar\omega_L)^{1/2}. \tag{B10f}$$

Solving (B10a,b) for $g_1(p_<)$ and $g_1(p_>)$ yields $f_1(\mathbf{p})$. The physical picture is little altered from the discussion of (B7a,b).