

Chapter 5

Dissipative electron-phonon system photoexcited far from equilibrium: beyond the Two-Temperature model

5.1 Abstract

The kinetics of evolution of the electron distribution function for an electron-phonon system driven far from equilibrium by photo-excitation is of considerable current interest, experimentally[69] (the pump-probe experiments) and theoretically[51, 46, 47] (e.g., the two-temperature model. In this Chapter[45], we have generalized a stochastic model known in the literature for the case of a driven dissipative granular gas[48]. We have derived the steady-state non-degenerate electron distribution for a semiconductor driven far from equilibrium by the inter-band photoexcitation assumed uniform over the nanoscale sample. Partitioning of the total inelastic electron scattering into the dissipative electron-electron and the electron bath(phonons) components is included in general. The model is applicable to a photoexcited semiconducting sample with fast removal of the electrons by electron-hole recombination from the bottom of the conduc-

tion band. The model is appropriate for a direct bandgap, disordered semiconducting system where the electron-hole recombination is fast, and the energy is the only state label.

5.2 The model.

Let $n_e(E)dE$ be the number of electrons lying in the energy range $\pm dE/2$ centred about E in the conduction band of a semiconducting sample of volume Ω . The electron-electron collisions, assumed inelastic in general, are described by the process; $E_i + E'_i \rightarrow E_f + E'_f = \alpha(E_i + E'_i)$ with $\alpha \leq 1$, in which the tagged electron of energy E_i collides with another electron of energy E'_i lying in the energy shell $E'_i \pm \frac{1}{2}\Delta E'_i$, and is scattered to the final state E_f . The scattering rate for this inelastic process is taken to be $(1 - f)\Gamma n(E'_i)dE'_i$. Similarly, the electron-phonon scattering rate is given by $f\Gamma n_{ph}(E'_i)dE'_i$, with $n_{ph}(E'_i)dE'_i$ as the number of thermal phonons in the phonon-energy shell $E'_i \pm \frac{1}{2}\Delta E'_i$. Here, the fraction $0 \leq f \leq 1$ determines the relative strengths of the binary electron-electron and the electron-phonon collisions. Also, let the electrons be injected through photo-excitation into the conduction band at energy E_{ex} at the rate $g_{ex}\delta(E - E_{ex})$, and then be removed (depleted) from the bottom of the conduction band through recombination. This depletion rate can be modelled by a term $-g_d\delta(E)n_e(0)$. Here the phonons are assumed to remain in thermal equilibrium at temperatures T . In our model sample we assume a uniform density of states for the electrons and the energy to be the only label for the single particle states. The photo-excitation is taken to be homogenous over the sample, which is reasonable for a nanoscale disordered semiconducting sample. For the above dissipative model driven far from equilibrium, the kinetics for the non-equilibrium electron number density $n_e(E)$ is given by the rate equation

$$\begin{aligned} \frac{\partial n_e(E)}{\partial t} = & -n_e(E) \int dE' [n_e(E')(1 - f) + n_{ph}(E')f]\Gamma \\ & + \int_0^1 dz p(z) \int dE' \int dE'' \delta(E - z\alpha(E' + E'')) n_e(E') n_e(E'') (1 - f)\Gamma \end{aligned}$$

$$\begin{aligned}
& + \int_0^1 dz p(z) \int dE' \int dE'' \delta(E - z(E' + E'')) n_e(E') n_{ph}(E'') f \Gamma \\
& + g_{ex}(t) \delta(E - E_{ex}) - g_d \delta(E) n_e(0).
\end{aligned} \tag{5.1}$$

In the above, we have assumed the total energy ($E' + E''$) for a binary collision to be partitioned such that a fraction z , with probability density $p(z)$, goes to the tagged electron of initial energy E' , and $1 - z$ to the colliding particle (electron or phonon of initial energy E''). The inclusion of a in the electron-electron collision takes care of the possibility of inelastic electron-electron collisions. Note that we have suppressed the time argument (t) in the non-equilibrium electron-number density $n_e(E)$. Taking the energy Laplace transform

$$\tilde{n}_e(s) = \int_0^\infty e^{-sE} n_e(E) dE, \tag{5.2}$$

of Eq. (1), we obtain,

$$\begin{aligned}
\frac{\partial}{\partial t} \tilde{n}(s) & = -\Gamma \tilde{n}_e(s) [(1-f)N_e + fN_{ph}] + (1-f)\Gamma \int_0^1 p(z) dz \tilde{n}_e^2(\alpha z s) \\
& + f\Gamma \int_0^1 dz p(z) \tilde{n}_e(zs) \tilde{n}_{ph}(zs) \\
& + g_{ex}(t) e^{-sE_{ex}} - g_d n_e(0).
\end{aligned} \tag{5.3}$$

In the following, we will consider for simplicity the steady-state condition under constant (cw) photoexcitation, $g_{ex}(t) = g_{ex}$. A pulsed excitation can, of course, be considered in general. Accordingly, we set $\frac{\partial}{\partial t} \tilde{n}_e(s) = 0$ above, and all quantities on the R.H.S. of Eq.(3) are then independent of time.

In order to calculate the steady-state electron distribution for the system in terms of the bath (phonon) temperature and other rate parameters, we expand $\tilde{n}_e(s)$ in powers of the Laplace-transform parameter s as

$$\tilde{n}_e(s) = N_e - s \langle E_e \rangle + s^2 \langle E_e^2 \rangle / 2 \dots, \tag{5.4}$$

and equate the co-efficients of like powers of s . Thus, from the zeroth power of s , we obtain at once

$$n_e(0) = (g_{ex}/g_d). \tag{5.5}$$

Similarly, from the first power of s , we get,

$$\langle e_e \rangle = \frac{(f/2)\langle e_{ph} \rangle}{\rho_{e-ph}(1-\alpha)(1-f) + f/2} + \frac{g_{ex}E_{ex}/\Gamma}{N_{ph}^2\rho_{e-ph}[\rho_{e-ph}(1-\alpha)(1-f) + f/2]}. \quad (5.6)$$

In the above, we have taken a uniform limit for the energy partition: $p(z) = 1$.

Here, we have defined $\langle e_e \rangle \equiv \langle E_e \rangle / N_e \equiv$ mean electron energy; $\langle e_{ph} \rangle \equiv \langle E_{ph} \rangle / N_{ph} \equiv$ mean phonon energy ($= k_B T_B$); and $\rho_{e-ph} = N_e / N_{ph} \equiv$ electron-to-phonon number ratio. It is to be noted that in the limit $a = 1$ (*i.e.*, for elastic electron-electron collisions as is usually expected for an electronic system unlike the case of the granular gas), and $g_{ex} = 0$ (*i.e.*, no photo-excitation), we recover $\langle e_e \rangle = \langle e_{ph} \rangle$, *i.e.*, the electrons and the phonons are at the same temperature, as is physically expected under equilibrium conditions. In general, however, the mean electron energy in the steady state is not the same as the mean phonon energy, and the former depends on the excitation rate (the drive g_{ex}).

We will now consider the specific case of the extreme partition limit $p(z) = \frac{1}{2}(\delta(z) + \delta(z-1))$ to illustrate our treatment. Analytic form for $\tilde{n}_e(s)$ in the steady state will be obtained for the case of elastic scattering ($a = 1$). It can be readily verified that for this insertion of $p(z)$ in Eq.(3), the Eqs. (5) and (6) remain unchanged. With this choice of $p(z)$, Eq.(3) now becomes

$$\begin{aligned} \tilde{n}_e(s)[(1-f)N_e + fN_{ph}] &= (1-f) \int_0^1 \left(\frac{1}{2}\delta(z) + \frac{1}{2}\delta(z-1) \right) dz \tilde{n}_e^2(zs) + f \int_0^1 \left(\frac{1}{2}\delta(z) \right. \\ &\quad \left. + \frac{1}{2}\delta(z-1) \right) dz \tilde{n}_e(zs) \tilde{n}_{ph}(zs) + g_{ex}[e^{-sE_{ex}} - 1]/\Gamma. \end{aligned} \quad (5.7)$$

This can be readily solved to give,

$$\tilde{n}_e(s) = \frac{2[(1-f)N_e + fN_{ph}] - f\tilde{n}_{ph}(s)}{2(1-f)} \pm \frac{\sqrt{[f\tilde{n}_{ph}(s) - 2[(1-f)N_e + fN_{ph}]]^2 - 4(1-f)C}}{2(1-f)} \quad (5.8)$$

with

$$C = (1-f)N_e^2 + fN_eN_{ph} + 2g_{ex}(e^{-sE_{ex}} - 1)/\Gamma.$$

We have to choose the $-ve$ sign in the above solution so as to satisfy $\tilde{n}_e(0) = N_e$, $\tilde{n}_{ph}(0) = N_{ph}$. Here, we have assumed that the bath phonons obey the equi-

librium Boltzmann distribution (*i.e.*, $\tilde{n}_{ph}(s) = N_{ph}/(1 + \langle E_{ph} \rangle s)$). Now, our boundary condition for the case of fast electron-hole recombination at the bottom of the band demands that the electron number density at the bottom of the conduction band be zero *i.e.*, $n_e(0) = 0$. But from the steady-state analysis, we have $g_{ex} = g_d n_e(0)$. Thus the above boundary condition is implemented mathematically by formally demanding that the drain co-efficient g_d tend to infinity, so that their product $g_d n_e(0)$ remains finite $= g_{ex}$. With this boundary condition, and using the initial-value theorem for Laplace transforms, we get a relation between the electron-to-phonon number ratio (ρ_{e-ph}) and the excitation parameter η as

$$\rho_{e-ph} = \frac{1}{2} \left[\sqrt{\beta^2 + \frac{8\eta}{1-f}} - \beta \right], \quad \eta = \frac{g_{ex}}{\Gamma N_{ph}^2}. \quad (5.9)$$

The electron number density distribution $n_e(E)$ (in the energy E-domain) is now obtained by numerically inverting $\tilde{n}_e(s)$ (in s-domain) in Eq(8), and is plotted in Fig(1) for a choice of electron-phonon coupling parameter f . (The noise seen on the lowest curve is an artifact of the numerical inversion).

At this stage, it is in order to comment briefly on our use of the extreme partition $P(z) = \frac{1}{2}\delta(z) + \frac{1}{2}\delta(1-z)$. First, let us note that our approach, based on Eq.(1), is of course valid for any general form of $p(z)$. It is, however, only for the above extreme limiting form that it admits an analytical solution as presented here in order to illustrate the model. The above form can now be justified in the following physical terms. It is to be noted that for any given collision, the partitioning of the total incoming energy between the out going particles necessarily depends on the kinematic parameters of the collision, *e.g.*, on the scattering angles, that vary from one collision to the other. Thus, for a fine-grained treatment of the successive collisions under the above extreme form, the energy of a 'tagged' electron colliding with others in the gas will fluctuate between extreme values. However, a coarse-graining over even a small number of successive collisions will suppress these fluctuations. Thus, in a coarse-grained statistical sense the partition is expected to become effectively a smooth and broad function. This

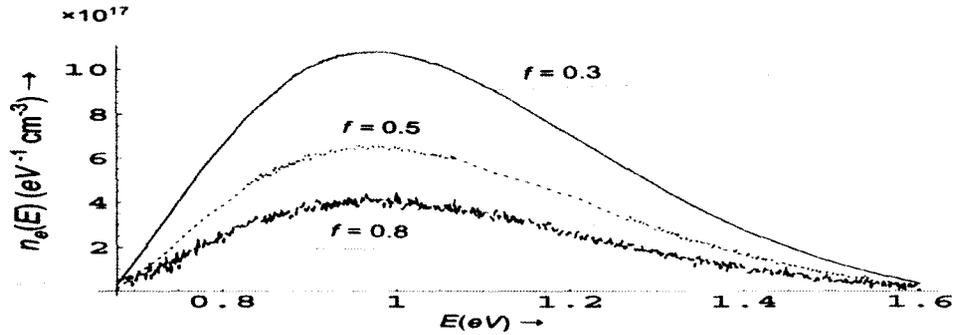


Figure 5.1: Plot of steady-state photoexcited electron distribution function $n_e(E)$ ($eV^{-1}cm^{-3}$) for a typical nanoscale semiconducting sample of radius $\sim 10nm$ with $\sim 10^5$ atoms, as function of the electron energy $E(eV)$, for the following choice of parameters: the excitation parameter $\eta = 10^{-7}$ and the electron-phonon coupling parameter $f = 0.3$ (*solid-line*), 0.5 (*dotted-line*), 0.8 (*lower-line*). In all cases, the peak occurs at $E = 1eV$ at which the electrons are being photo-injected in the conduction band. Here the phonons are assumed to remain at the room temperature ($300K$).

physically justifies our use of the simple, though admittedly extreme, form for the partition $p(z)$ without resulting in unphysical features, as validated a posteriori from Figure 6.1.

5.3 Discussion.

We have treated here the problem of energy-distribution of photo excited electrons in a semiconducting sample as a generalization of the model for dissipative granular gas driven far from equilibrium. A feature of the above non-equilibrium distribution of the cw photo-excited electrons is the peak appearing at the excitation energy(= $1eV$) as is indeed expected. However, a notable feature of the steady-state electron distribution in this case is that, as the electron-phonon interaction strength (f) increases, (1) the peak height of the distribution decreases, as also (2) the total area under the distribution (the total number of steady-state photoexcited electrons in the conduction band). This is physically understandable as follows. Inasmuch as the increased

electron-phonon interaction implies fast removal of energy from the non-equilibrium photo-excited electron distribution to the phonon bath, the electrons get pushed to lower energies towards the bottom of the conduction band, from where they get removed by the fast electron-hole recombination. Hence the decrease in the height of the distribution, and also reduction of the area under the curve. It should be possible to probe these steady-state features through a cw pump-probe experiment. As for our assumption of non-degeneracy for the photoexcited electron gas, it is readily verified that the peak occupation number of the one-particle states is of the order of $(N_e/\text{total number of atoms in the nanoscale sample}) \sim 10^{-4} \ll 1$, thus validating the assumed non-degeneracy. Finally, for the case of dissipative electron-electron collisions ($a < 1$), it can physically derive from the dissipative polarization of the dielectric medium that screens the electron-electron Coulombic interaction[49].