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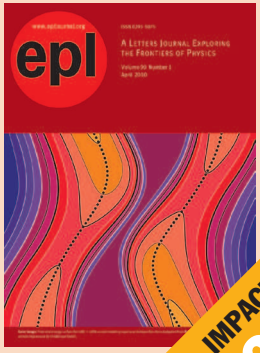

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Universal anomalous exponent of photoconduction

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Abstract – Experiments often show that the photoconductance σ of a semiconductor system and the light intensity I are related by $\sigma \sim I^\gamma$. Conventional theories give a satisfactory explanation for $\gamma = 1$ or $\gamma = \frac{1}{2}$, but anamalous exponents close to $\gamma = \frac{3}{4}$ are often observed. This paper argues that there is a universal anomalous regime for which $\gamma = \frac{3}{4}$ (or $\gamma = \frac{2}{3}$ in two dimensions), resulting from the kinetics of electron-hole recombination being controlled by Coulombic attraction. Because the local electric fields are extremely high, the theory uses the "hot-carrier" model for transport.

Introduction. – Many semiconductor devices rely on excess charge carriers which are either injected or else created by absorbing photons. Electron-hole recombination is a process which is central to undestanding the dynamics of these systems. The radiative and non-radiative mechanisms for recombination have been extensively studied, starting with seminal work of Shockley and Read [1]. Here I argue that there is a universal regime for the kinetics of recombination which has not been described in earlier work. The evidence comes from considering an anomalous exponent describing the response of photoconductors, where the photoconductance σ and the light intensity I are typically related by $\sigma \sim I^\gamma$: values close to $\gamma = \frac{3}{4}$ are frequently observed, whereas the law of mass action only explains $\gamma = 1$ or $\gamma = \frac{1}{2}$. I argue that accounting for the role of Coulomb interaction in the kinetics of electron-hole recombination leads to an explanation for values of the anomalous exponent, γ. Because the local electric fields between the carriers are extremely high, the theory uses the "hot-carrier" model for transport, where the drift velocity becomes nearly independent of the field [2,3].

Consider a photocell using an undoped semiconductor element. Light falling on the device creates electron-hole pairs (at a rate which is proportional to the intensity of radiation, I). If the density of mobile electrons is n_e and the density of holes is n_h, the conductivity is $\sigma = e(\mu_\mathrm{e} n_\mathrm{e} + \mu_\mathrm{h} n_\mathrm{h})$, where μ_e and μ_h are the electron and hole mobilities, and e is the electron charge. Because the semiconductor is intrinsic, $n_\mathrm{e} = n_\mathrm{h} \equiv n$. The rate of change of the excitation density n has a source term, proportional to the intensity, and a sink term, which represents the rate for electron-hole recombination: $\mathrm{d}n/\mathrm{d}t = AI - f(n)$ where A is a constant and $f(n)$ is the rate for recombination of electron-hole pairs at density n. It is natural to invoke the law of mass action, and to propose that the rate of recombination is $\mathrm{d}n_\mathrm{e}/\mathrm{d}t = \mathrm{d}n_\mathrm{h}/\mathrm{d}t = -Cn_\mathrm{e}n_\mathrm{h}$ (for some constant C), that is $f(n) = Cn^2$. In this case the equilibrium concentration n and consequently the conductance are proportional to $\sqrt{I}$. An alternative scenario is that electrons and holes become immobilised on "recombination centres", which exist independently of the illumination: this leads to a prediction that $\sigma \propto I$. Thus there appear to be two plausible models for the recombination rate, which are distinguished by looking at the exponent γ for a power-law dependence of conductance G upon light intensity:

$$G \sim I^\gamma. \tag{1}$$

Both $\gamma = 1$ and $\gamma = \frac{1}{2}$ are observed in different experiments, corresponding, respectively, to mobile charge carriers being destroyed by "monomolecular" or "bimolecular" processes. But a surprising aspect of the literature is that intermediate values of γ (hereafter termed *anomalous*) are often observed over several decades of intensity.

A theory for the anomalous exponents has been discussed by Rose [4], but his model is unsatisfying because it assumes that there is a set of impurity states in the band gap with a density of states which is an exponential function of energy. Studies of Lifshitz tails indicate that the density of states has a more complex functional dependence: see [5,6]. The theory in [4] also

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Table 1: Summary of previously published anomalous exponents, γ: "N decades" means "observed over N decades of intensity".

Reference	Material	γ	Remarks
Arene and Baixeras [8]	a-Si H	0.78	4 decades
Arene and Baixeras [8]	a-Si H	0.55	3.5 decades
Bakr [9]	As_2Sb_3 film	0.58–0.71	1.5 decades
Jie *et al.* [10]	Cd S nanowires	0.74–0.77	1.5 decades
Kaplan and Kaplan [11]	As_2Se_3	0.44–0.86	2 decades, ω varied
Kaplan and Kaplan [12]	a-Se	0.61–0.96	2 decades, ω varied
Kind *et al.* [13]	Zn O nanowires	0.8	4 decades
Rose [4]	Sb_2S_3 (Vidicon cameras)	0.68	"several decades"
Spear *et al.* [14]	a-Si	0.54–0.9	method unclear
Wronski and Daniel [7]	a-Si H	0.83	2-3 decades, T varied
Wronski and Daniel [7]	a-Si H	0.7	3 decades

indicates that γ should vary with temperature, T. This is not compatible with observations in [7] where γ was found to be independent of T (consistent with the "universality" hypothesis developed here).

Below I argue that where anomalous exponents are reported, they are frequently close to $\frac{3}{4}$, particularly in cases where a good fit to a power-law is demonstrated over a wide intensity range. This suggests that $\gamma = \frac{3}{4}$ could be a "universal" exponent for the anomalous regime. The samples in these experiments often require specialist preparation, and the reported values of γ are not always consistent. In view of the very surprising nature of the anomalous photoconductive response, it is desirable to characterise the effect in a readily accessible system. With this motivation I also measured the response of four commercially available photocells. An anomalous regime with $\gamma = \frac{3}{4}$ was observed in three of the samples, and $\gamma = \frac{2}{3}$ was observed in the fourth. I conclude by presenting a theory explaining these universal exponents.

Discussion of earlier work. – Anomalous photoconductive response has been observed in a wide variety of semiconductors and devices. The exponents which I have found reported are listed in table 1. Some authors [9,11,12,14] report a broad range of values of γ, depending upon temperature or other parameters. In the remaining cases where either a single value of γ or a narrow range is reported, its value lies in the range 0.7–0.8, apart from three "outliers", discussed below. Small deviations of the exponent fitted in experiments can result from various effects, such as the limits of the intensity range extending into regions where the anomalous regime is breaking down, or the properties of the materials being modified by the concentration of mobile carriers. For this reason, it is unlikely that differences in these values of γ are significant. The literature is consistent with the existence of a robust regime with an exponent close to 0.75, while different exponents may be observable in other systems.

There are three "outliers", where a single value of γ is quoted which is not in the range 0.7–0.8. Wronski and Daniel [7] show results which are consistent with $\gamma = 0.83$ over a range of temperatures. Rose [4] reports that Vidicon camera tubes (which used Sb_2S_3) give $\gamma = 0.68$ over "several decades". Arene and Baixeras [8] show a plot with $\gamma = 0.55$ over nearly four decades, but the exponent is so close to $\gamma = \frac{1}{2}$ that it is questionable whether this is an anomalous response.

Rose [4] predicts that γ depends upon the temperature T: $\gamma = T_0/(T + T_0)$ for $T \leqslant T_0$, where T_0 is an effective temperature characterising the exponential density of states in the band gap. Wronski and Daniel [7] examined the intensity dependence of the photocurrent in hydrogenated amorphous silicon at a variety of temperatures ranging from 147 K to 267 K. Figure 5 of their paper demonstrates that the exponent γ is close to 0.83 and independent of T at lower intensities, crossing over to 0.5 at higher intensity.

On the other hand several papers [9,11,12,14] report that γ does depend upon temperature (or upon modulation frequency, ω) but their exponents vary erratically. Table 1 does not include reports (such as [15,16]) where the samples displayed non-Ohmic behaviour.

Investigation on commercial photocells. – I have pointed out that much of the the published experimental data are consistent with a previously un-remarked universality, where $\gamma \approx \frac{3}{4}$. Some of the other literature suggests, however, that the values of the anomalous exponent are highly variable and may be erratic and hard to reproduce. By definition, universal behaviour does not shy away from observation. It might be observable in a simple experiment with a generic semiconductor sample. With this motivation, I measured the response of four different commercially available light-dependent resistors at room temperature. The photocells were obtained from Maplin Electronics: their catalogue numbers were: sample 1 - N56AY, sample 2 - N57AY, sample 3 - N53AY, sample 4 - N46AY. The intensity of light from a tungsten bulb was reduced by stopping down the aperture of a light box, and by increasing the distance between the aperture and the photocell. Some of the higher intensity data used an unshielded 15 W bulb, varying the distance from the

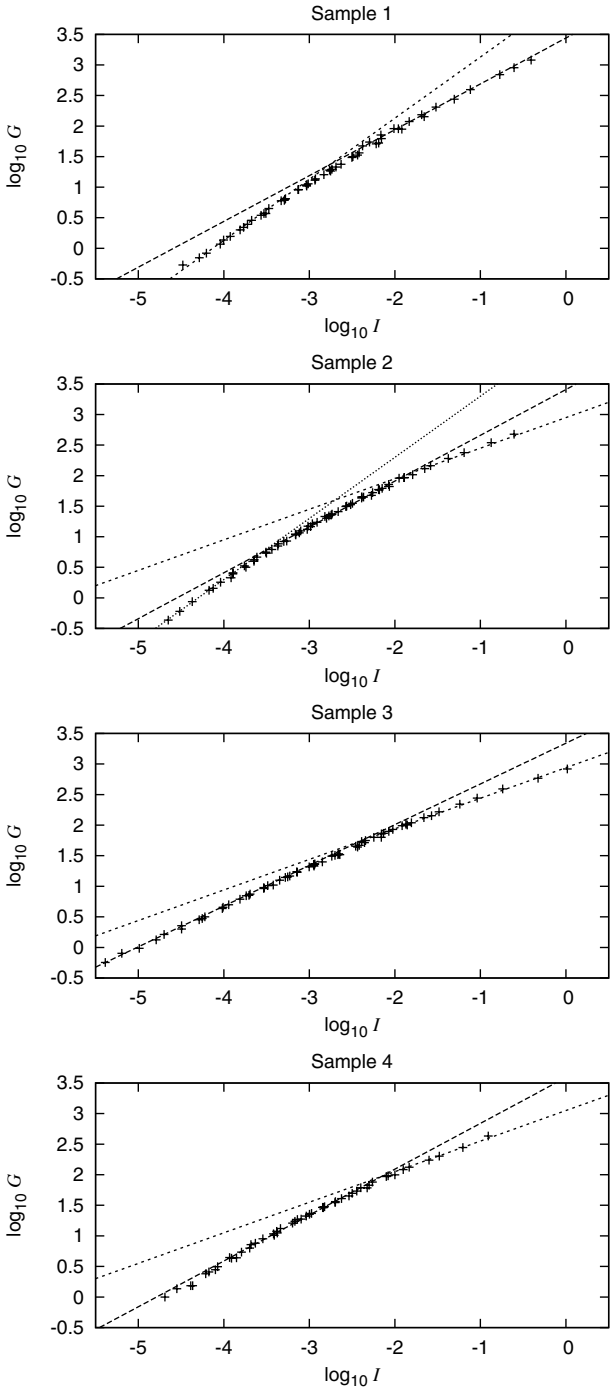

Fig. 1: Plot of $\log_{10} G$ (the conductance G measured in units of $10^{-6}\Omega^{-1}$) *vs.* $\log_{10} I$ (with the intensity I in arbitrary units). Each sample showed an anomalous regime. The slopes of the linear fits are as follows. Sample 1: $\gamma = 1$, $\frac{3}{4}$, Sample 2: $\gamma = 1$, $\frac{3}{4}$, $\frac{1}{2}$, Sample 3: $\gamma = \frac{2}{3}$, $\frac{1}{2}$, Sample 4: $\gamma = \frac{3}{4}$, $\frac{1}{2}$.

sample. The response of each photocell was found to be Ohmic for both low and high light intensities, and its dark current was negligible. Figure 1 shows logarithmic plots of conductance G (in units of $10^{-6}\Omega^{-1}$) *vs.* intensity I. The light intensity scale is in non-standardised units: the 15 W pearlescent bulb at 1 m gave an intensity $I = 4 \times 10^{-3}$. For each sample there was clear evidence for an anomalous regime. Sample 1 shows a monomolecular regime ($\gamma = 1$) at low intensity, crossing over to an anomalous regime at higher intensity for which $\gamma = \frac{3}{4}$ gives an excellent fit over two decades. Sample 2 showed an anomalous regime with $\gamma = \frac{3}{4}$ spanning nearly two decades at intermediate intensities between regimes with $\gamma = 1$ and $\gamma = \frac{1}{2}$. Sample 3 showed an anomalous regime for which $\gamma = \frac{2}{3}$ provides an excellent fit over three decades, crossing over to bimolecular behaviour ($\gamma = \frac{1}{2}$) at high intensities. Sample 4 shows an anomalous regime for which $\gamma = \frac{3}{4}$ provides a good fit, crossing over to $\gamma = \frac{1}{2}$ at high intensity.

Three out of four samples showed anomalous behaviour with $\gamma = \frac{3}{4}$, providing strong support for the hypothesis that this is a "universal" phenomenon. The other sample produced an exponent very close to that reported by Rose [4] for the Vidicon camera.

Theory for anomalous exponent. – There are as many as three different regimes which can be observed at different light intensities. At the lowest intensities a carrier is likely to encounter a trap before meeting a carrier with the opposite sign, and there is a *monomolecular regime*, where $\gamma = 1$. At intermediate intensities, there may be an *anomalous regime*, the origin of which is considered below. At the highest light intensities, carriers are removed by electron-hole recombination at a rate proportional to the product of the electron and hole concentrations. In this *bimolecular regime*, $\gamma = \frac{1}{2}$.

As the terms *monomolecular* and *bimolecular* suggest, the exponents $\gamma = 1$ and $\gamma = \frac{1}{2}$ are both explained by invoking the law of mass action, as outlined in the introduction. The law of mass action is applicable in situations where the reactive species move randomly by diffusion, without any interaction occurring until they are in contact. This is realistic for chemical reactions in which there is no electrostatic interaction. In the case of electron-hole recombination, Coulombic attraction between electrons and holes can pull them together, so that the time required for recombination may be less than it would be for randomly diffusing particles. The following discussion uses scaling arguments rather than detailed calculations of the electron dynamics, but these arguments are sufficient to make a precise determination of the anomalous exponent.

The effect of recombination events on the density n may be written $\mathrm{d}n/\mathrm{d}t = -n/\tau$, where n is the number of either species, and τ is the lifetime for an electron or hole to survive without collisions. If the collisions occur due to random diffusion of electrons and holes, then the lifetime τ will be inversely proportional to the density of the other species, so that $\mathrm{d}n/\mathrm{d}t \propto n^2$. If the Coulombic force is significant, the timescale for collision will be the time required for an electron and a hole to be pulled together by their mutual electrostatic attraction, and τ is expected to be proportional to the characteristic distance L that an electron and a hole have to travel in order to make

a collision. The timescale is also inversely proportional to the characteristic velocity v for relative motion of the electrons and the holes: $\tau \sim L/v$. If the density of carriers is n, we clearly have $L \sim n^{-1/d}$, where d is the effective spatial dimension of the photosensitive region penetrated by the radiation (that is, $d=3$, unless the depth is small compared to L, when we would use $d=2$). The velocity v will depend upon the local electric field $\mathcal{E}$ driving the drift towards collision. The charges $\pm e$ of the electrons and holes are independent of the density, but the lengthscale L depends upon the carrier density so that $\mathcal{E} \sim L^{1-d} \sim n^{(d-1)/d}$.

In order to determine how τ depends upon n, it remains to specify how the typical drift velocity v depends upon the electric field $\mathcal{E}$. At first sight it seems natural to invoke linear response theory, and to propose that $v \propto \mathcal{E}$. However, because the lengthscale L is very small, the electric field $\mathcal{E}$ will be very large. For example, the electric field due to an electron at a distance of $10^{-8}\,\mathrm{m}$ is of order $10^{7}\,\mathrm{V\,m^{-1}}$. Also, the electrostatic attraction between an electron and the closest hole is not screened, because the space between them contains no mobile charges. The electrostatic fields are, therefore, so high that the applicability of linear response theory is questionable. It is argued that "hot carriers" in semiconductors have a damping rate due to interaction with phonons which rises very steeply with energy [2,3]. As a consequence, the velocity of mobile carriers in semiconductors becomes insensitive to the applied electric field, and may even approach a limiting value as the electric field increases [17,18]. Hot-carrier effects are typically considered to be significant for electric fields in excess of $10^{5}\,\mathrm{V\,m^{-1}}$ [2,3].

The physics of the hot-carrier model will be discussed in greater detail below, after considering its consequences. It will be assumed that the relation between carrier velocity and electric field may be described by a power-law, such that

$$v \sim \mathcal{E}^{\beta}. \tag{2}$$

Linear response theory corresponds to setting $\beta=1$. The hot-carrier model, where the velocity is quite insensitive to the microscopic electric field, is consistent with either assigning a very small value to β, or else taking the limit as $\beta \to 0$. Before discussing the value of β, consider the formula for the anomalous exponent. The recombination rate is

$$\mathrm{d}n/\mathrm{d}t \sim -nv/L \sim nn^{1/d}\mathcal{E}^{\beta} \sim n^{\left(1+\frac{1}{d}+\frac{\beta(d-1)}{d}\right)}. \tag{3}$$

Because the rate of recombination is proportional to the flux I, $n \sim I^{\gamma}$ with

$$\gamma = \frac{d}{d+1+(d-1)\beta}. \tag{4}$$

Equation (4) is the central theoretical result of this paper. If the hot-carrier theory is applied and the limit $\beta \to 0$ is taken, the exponent is $\gamma = \frac{3}{4}$ in three dimensions. For $d=2$, the hot-carrier model gives $\gamma = \frac{2}{3}$, which is consistent with the results for sample 3, with the exponent quoted by Rose [4] for Vidicon cameras, and with the mid-range for the results of Bakr [9] on As_2Sb_3 films.

Other authors have suggested that Coulomb interactions influence the rate of electron-hole recombination, presenting theories which imply a different value for the exponent γ. Langevin considered the influence of Coulombic attraction on the recombination of ions in discharge tubes (see [19] for a discussion of this early literature). His work assumes that the drift velocity towards collision is determined by the mobility of the carriers, which implies setting $\beta=1$ in (4). Langevin's approach has been adapted to recombination in semiconductors (see, for example [20]), but this approach leads to the exponent $\gamma=1/2$ in three-dimensions, the same as for bi-molecular recombination, and does not explain the anomalous photoconductive response. Other authors (for example, [21,22]) have considered electron-hole correlations which modify the constant C in the mass action formula for the collision rate, $\mathrm{d}n_{\mathrm{e}}/\mathrm{d}t = C\,n_{\mathrm{e}}n_{\mathrm{h}}$. Their calculations are only relevant to the bi-molecular process.

Discussion of the hot-carrier model. – Experimentally, it is found that the drift velocity of carriers in semiconductors increases very slowly with the electric field beyond approximately $10^{5}\,\mathrm{V\,m^{-1}}$ [2]. It has been suggested that the drift velocity may saturate at approximately $(\hbar\omega_0/m)^{1/2}$, where ω_0 is optical phonon frequency and m the effective carrier mass [2]. This is consistent with setting $\beta=0$ in (4), but the argument supporting this suggestion was not made clear in [2]. Most theoretical works on the hot-carrier picture are concerned with numerical modelling for specific systems (see, for example, [17,18]). In the following I discuss a simplified but plausible model which justifies choosing a small but non-zero value for the exponent β in (2).

Because we consider the microscopic structure of the electric field on a scale of the separation of the carriers, the field is not screened by polarisation effects. The samples where the anomalous response is observed are usually amorphous or highly disordered microcrystalline systems. The following discussion will assume that the carriers and the phonons are moving in a highly disordered environment.

The electric field $\mathcal{E}$ will cause an electron (or hole) to accelerate to a velocity v corresponding to an energy $E=\frac{1}{2}mv^2$, where m is the effective mass of the carrier. A particle with velocity v will then gain energy from the electric field at a rate $\dot{E} = e\mathcal{E}v \sim \mathcal{E}E^{1/2}$. This acceleration is opposed by the tendency of the motion of the charged particle to excite phonons: a particle with energy E will dissipate energy to phonons at a rate which will be written $\dot{E} = R(E)E$, where $R(E)$ is a rate coefficient. The energy of a carrier accelerated by a constant electric field is therefore given by the solution of the energy balance

equation

$$\frac{\mathrm{d}E}{\mathrm{d}t} = e\mathcal{E}\sqrt{2E/m} - R(E)E. \tag{5}$$

The hot-carrier picture asserts that rate coefficient $R(E)$ increases very rapidly with energy E. Various arguments and calculations have been advanced to support this claim [3,17,18]. In the following I present a simplified model which yields $R(E) \sim E^{9/2}$.

The loss of energy by an electron to the phonons can be seen as analogous to spontaneous emission from an atomic state, with the phonons playing a role analogous to the photons in electrodynamics. Recall the expression for the rate R_{nm} of spontaneous emission from an atomic level with energy E_n to a level with lower energy, E_m: $R_{nm} = (4\alpha/3c^2)|M_{nm}|^2\omega_{nm}^3$, where $\omega_{nm} = (E_n - E_m)/\hbar$ is the frequency of the photon which is emitted, and M_{nm} is the corresponding dipole matrix element (c is the speed of light and α is the fine-structure constant). This expression can be adapted to describe the spontaneous decay of electronic states by emission of phonons. For simplicity it will be assumed that the matrix elements M_{nm} for the electron-phonon coupling in the highly disordered environment are random numbers, with statistics which are independent of the energies E_n and E_m. The rate of emission into any given state of lower energy is therefore proportional to ΔE^3, where $\Delta E = E_n - E_m$. The number of available electronic states with energy below E is $N(E) \sim E^{3/2}$. Upon integrating over the lower energy, the rate coefficient scales as $R(E) \sim E^{9/2}$. The steady-state solution of equation (5) is therefore $\mathcal{E} \sim E^5$, so that the terminal velocity is $v \sim \mathcal{E}^{1/10}$, that is $\beta = \frac{1}{10}$. This supports the assertion of the hot-carrier model, that the electron or hole drift velocity becomes highly insensitive to the electric field. Some authors (for example, [2,17,18]) have gone further, suggesting that the drift velocity approaches an asymptote as the electric field increases.

Conclusions. – I have presented evidence that the anomalous exponent of photoconduction is universal, with $\gamma = \frac{3}{4}$ for $d = 3$, $\gamma = \frac{2}{3}$ for $d = 2$. This is explained by a breakdown of the mass action principle, with the kinetics of electron-hole recombination being driven by Coulomb attraction. Because the electric fields are extremely high the theory invokes the hot-carrier transport model.

* * *

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