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Verification of a generalised Planck law by investigation of the emission from GaAs luminescent diodes

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Abstract. A generalisation of Planck's law for the spontaneous emission from light emitting diodes with direct band gaps allows a calculation of the absolute spectral emission intensity to be performed when the applied voltage, temperature, and absorption coefficient of the light emitting region are known. For given values of these parameters the emission intensity is independent of the diode current and the quantum efficiency of the recombination process. GaAs LED spectra calculated on this basis are compared with measurements at 82 K and at 296 K, and are found to agree quite well. Differences are attributed to the uncertain knowledge of the absorption coefficient of the diode material.

1. Introduction

The usual approach for calculating the rate of radiative recombination in semiconductors based on the work of van Roosbroeck and Shockley [1] is an approximation. For a semiconductor without external excitation or any applied voltage it does not (as it should) yield Planck's emission formula. The deviation, however, is negligible as long as the energy of the emitted photons is large compared with kT and as long as the emission is far from laser emission.

This deficiency has been corrected by Lasher and Stern [2]. The emission from semiconductors up to the lasing threshold can then be described exactly, even under 'non-equilibrium' conditions, i.e. with external excitation or applied voltage, by introducing into Planck's law the chemical potential, μ_γ , of photons [3], which is equal to the difference of the quasi-Fermi energies as long as these govern the distribution of electrons and holes in their respective bands.

As a result of the thermodynamical treatment in [3] the photon emission from LEDs with direct band-band transitions is recognised as being in chemical equilibrium with the electron-hole distribution and is therefore fully determined by the absorption coefficient of the diode material, the temperature of the diode, and the voltage applied to it, which causes the quasi-Fermi energies to be different for electrons and holes. This result is independent of the presence of non-radiative transitions. Other quantities usually related to the emission from LEDs, e.g. the diode current or the quantum

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efficiency, turn out to be irrelevant. In simple terms: for a given voltage all diodes made from the same material in the same geometry should emit the same intensity. Whether they are good or bad shows up in the diode current, which is larger for bad, less efficient diodes since it must provide additional electrons and holes for non-radiative recombination.

Due to the prevailing treatment of photon emission by rate equations, its simple dependence on the difference of the quasi-Fermi energies has obviously been overlooked. In the large number of publications concerned with light emission from GaAs diodes no comprehensive set of data on the absolute spectral emission intensity could be found to test the predicted voltage dependence. In the following we report on measurements of the emitted absolute spectral intensity from GaAs diodes and compare them with theoretical expectations.

2. Theory of diode emission

Our thermodynamical approach takes advantage of the existence of thermal equilibrium, i.e. all particles involved, electrons e , holes h , phonons Γ , and photons γ of the emitted radiation have the same temperature, and their distribution over energy is given by Fermi and Bose–Einstein distribution functions, respectively. It also takes advantage of the existence of chemical equilibrium of the reaction



which leads to

$$\eta_e + \eta_h = \mu_\gamma. \quad (2)$$

This chemical equilibrium is the consequence of the large rate of reabsorption in the diode due to the large value of the absorption coefficient, and because most photons cannot leave the crystal due to the high index of refraction of GaAs ($n = 3.4$).

The sum of the electrochemical potentials of electrons and holes $\eta_e + \eta_h$ may be better known as the difference of the respective quasi-Fermi energies $\varepsilon_{F,e}$ and $\varepsilon_{F,h}$. Thus

$$\mu_\gamma = \varepsilon_{F,e} - \varepsilon_{F,h}. \quad (3)$$

This relation can be tested in pn junctions with applied voltage U , where

$$\varepsilon_{F,e} - \varepsilon_{F,h} = eU \quad (4)$$

if the voltage drop across the series resistance is neglected.

In the following we consider a pn junction in the geometry of figure 1. The light generated in the n region of the GaAs diode is neglected because of the small diffusion length of holes into the n region compared with the diffusion length of electrons into the p region and because the light from the n region has to pass the p region, where it is strongly absorbed before it can leave the diode. As shown in figure 1 the difference of the quasi-Fermi energies $\varepsilon_{F,e} - \varepsilon_{F,h}$ is equal to eU at the pn junction, from where it decreases into the p region by kT over a diffusion length L_e , falling to zero at the end of the p region of width d , i.e. at the surface, due to the large surface recombination velocity. Taking the electron current as a pure diffusion current, i.e. neglecting any field

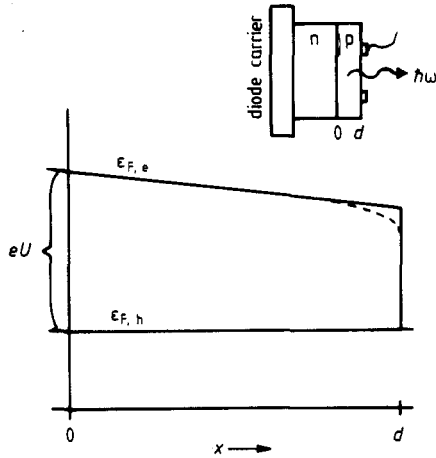


Figure 1. The spatial variation of the quasi-Fermi energies, $\epsilon_{F,e}$ for electrons and $\epsilon_{F,h}$ for holes, in the $15\ \mu\text{m}$ thick p side of the GaAs LED corresponding to the electron distribution in equation (5a) (broken curve) and in equation (5b) (full curve).

contribution from a space charge layer at the surface, the spatial variation of the electron density n_e is [4]

$$n_e(x) = n_e(0) \sinh[(d - x)/L_e] / \sinh(d/L_e). \tag{5a}$$

Although it is likely that there is a space charge layer, it is rarely taken into consideration, since its effect on the electron distribution is not analytically treatable. We have solved the continuity equation for the electron current in the presence of a space charge layer numerically. We have found that the electron distribution is well represented by equation (5a) if holes are depleted in a surface space charge layer. If the space charge layer is a hole accumulation layer, the electron density n_e falls off much more rapidly at the surface. As a result n_e is larger than in equation (5a) just outside the space charge layer, which is less than $0.1\ \mu\text{m}$ wide, and is better represented by a distribution for diffusion into a p side of infinite extension:

$$n_e(x) = n_e(0) \exp(-x/L_e) \tag{5b}$$

with a discontinuous drop at the surface.

Equations (5a) and (5b) are regarded as extreme cases. The actual distribution may be closer to (5b) because the surface region is more highly doped to ensure good lateral conduction.

The continuity equation for a photon current of density j_γ propagating into the x direction is

$$(d/dx)j_\gamma(x) = (1/4n^2)r_{sp}(x) - (r_a(x) - r_{st}(x)) \tag{6}$$

where r_{sp} , r_a and r_{st} are the rates per volume of spontaneous emission, absorption and stimulated emission, respectively. According to Lambert's law a quarter of the photons emitted into 4π inside the diode would be able to pass through the surface if the refractive index n were the same on both sides of the surface. Since the density of states for photons is larger in GaAs than outside by a factor of n^2 , only the fraction $1/4n^2$ of all emitted photons are not totally reflected. They are emitted through the front surface if it is ideally anti-reflection coated. The fraction $1/4n^2$ of all photons fills only a small solid angle, which justifies the one-dimensional approach in equation (6).

The difference $r_a - r_{st}$ is the net absorption rate and is related to the absorption coefficient α by

$$r_a - r_{st} = \alpha(\hbar\omega)j_\gamma. \quad (7)$$

Equations (6) and (7) lead to the differential equation

$$(d/dx)j_\gamma(x) = (1/4n^2)r_{sp}(x) - \alpha(\hbar\omega, x)j_\gamma(x). \quad (8)$$

With $j_\gamma(0) = 0$ its general solution is

$$j_\gamma(x) = \frac{1}{4n^2} \exp\left(-\int_0^x \alpha(\hbar\omega, x') dx'\right) \int_0^x r_{sp}(x') \exp\left(\int_0^x \alpha(\hbar\omega, x') dx'\right) dx'. \quad (8a)$$

Neglecting dispersion the rate of spontaneous emission into a solid angle of 4π is from (3):

$$r_{sp}(x) = \alpha(\hbar\omega, x) \frac{n^2(\hbar\omega)^2}{\pi^2 \hbar^3 c^2} \left[\exp\left(\frac{\hbar\omega - \mu_\gamma(x)}{kT}\right) - 1 \right]^{-1}. \quad (9)$$

Except for the chemical potential μ_γ of the emitted photons this expression is identical to Planck's expression for the emission rate in a black body. For all cases of interest $\hbar\omega - \mu \gg kT$ and the -1 in the denominator of (9) can be neglected.

The spatial distribution of the absorption coefficient $\alpha(\hbar\omega, x)$ depends on the distribution of the doping concentration, which is not exactly known. Taking it as spatially constant is a good approximation at room temperature, where all of the p region of width d contributes to the photon emission. With $\mu_\gamma(x)$ given by

$$\mu_\gamma(x) = eU + kT \ln(n_c(x)/n_c(0)) \quad (10)$$

the integration of (8a) for the distribution in (5a) is

$$j_\gamma(d) = \frac{(\hbar\omega)^2}{4\pi^2 \hbar^3 c^2} \frac{\exp[(eU - \hbar\omega)/kT]}{2 \sinh(d/L_e)} \left(\frac{\alpha}{\alpha - 1/L_e} \{1 - \exp[-(\alpha - 1/L_e)d]\} - \frac{\alpha}{\alpha + 1/L_e} \{1 - \exp[-(\alpha + 1/L_e)d]\} \right). \quad (11a)$$

The integration of equation (8a) for the distribution in (5b) is

$$j_\gamma(d) = \frac{(\hbar\omega)^2}{4\pi^2 \hbar^3 c^2} \frac{\exp[(eU - \hbar\omega)/kT]}{\exp(d/L_e)} \frac{\alpha}{\alpha - 1/L_e} \{1 - \exp[-(\alpha - 1/L_e)d]\}. \quad (11b)$$

Finally, the energy current density j_E emitted by the diode through the front surface with reflectance $R(\hbar\omega)$ into a solid angle of 2π is

$$j_E(\hbar\omega) = (1 - R(\hbar\omega))\hbar\omega j_\gamma(d). \quad (12)$$

Equations (11) and (12) show that the absolute spectral intensity is fully determined by the temperature T , the absorption coefficient α and the voltage U applied to the diode.

A formalism as described above has been used to extract values of the diffusion length L_e from relative emission data [5].

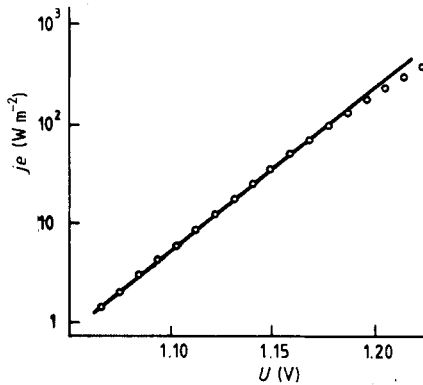


Figure 2. The experimental (circles) voltage dependence of the total emission intensity into 2π at $T = 296$ K.

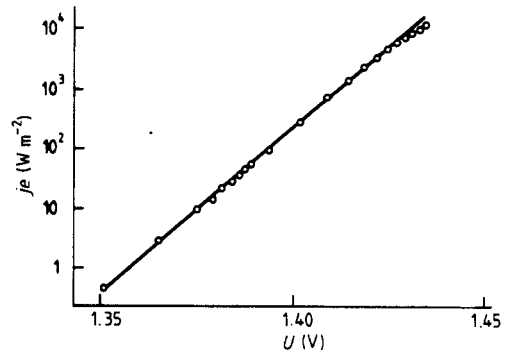


Figure 3. The experimental (circles) voltage dependence of the total emission intensity into 2π at $T = 82$ K.

3. Experimental results

The diodes used for the measurements are GaAs light emitting diodes (n region: $(5-10) \times 10^{17} \text{ cm}^{-3}$ Si; p region: $3 \times 10^{18} \text{ cm}^{-3}$ Zn) fabricated by diffusing Zn into GaAs:Si base material. The pn junction is located $15 \mu\text{m}$ behind the front surface, which is anti-reflection coated with Si_3N_4 . The diode chips are mounted with the n side down on a metal carrier as shown in figure 1.

The spectral intensity was measured with a calibrated Si pin-diode behind a grating monochromator. Absolute values were calculated by matching the integrated spectral intensity to the total intensity measured without the monochromator.

3.1. Voltage dependence

The emission formula (12) predicts that the emission intensity increases exponentially with the applied voltage. This dependence is confirmed by the experimental results shown in figures 2 and 3 for $T = 296$ K and 82 K, respectively. The deviation at the higher voltages is due to the voltage drop across the series resistance caused by the large electric current.

3.2. Absolute spectral intensity

Our diodes' spectral emission intensity per solid angle was found to vary with the angle to the surface normal according to Lambert's law, as was found previously [6, 7]. This is the expected result for the large value of the absorption coefficient. A single measurement of the angular dependence at one voltage was therefore sufficient to calculate the intensity emitted into 2π from the intensity per solid angle perpendicular to the diode surface.

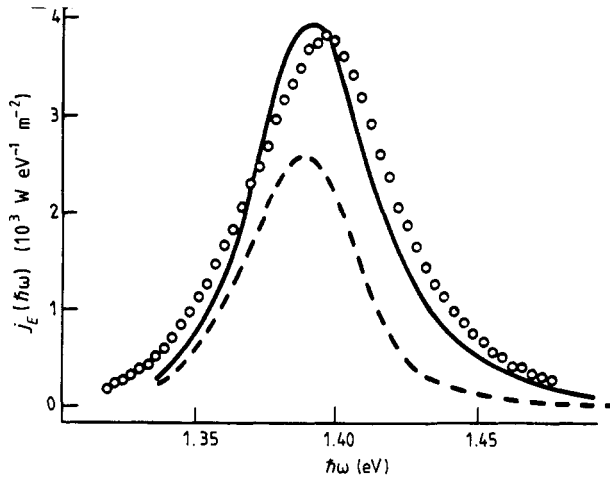


Figure 4. The experimental (circles) and theoretical (curves) absolute spectral energy current density emitted by a LED into 2π as a function of photon energy $\hbar\omega$ at $T = 296$ K and a voltage of 1.1206 V. The broken curve is based on the spatial distribution of electrons in equation (5a) and the full curve is based on the distribution in equation (5b).

3.2.1. Room temperature. The measured spectral intensity at $T = 296$ K is shown in figure 4. It is compared with the theoretical absolute spectral intensity as calculated from (12) for the two electron distributions in (5a) and (5b). The diffusion length L_e at room temperature was found to be equal to the width of the p region from the observation of the side emission of the diode under a microscope with the help of a CCD camera. The absorption coefficients used for the calculations of (12) at $T = 296$ K are taken from [8] for $\hbar\omega \leq 1.39$ eV and from [9] for $\hbar\omega \geq 1.39$ eV. These data relate to GaAs with doping concentrations slightly different from those in the material of our diodes. The Si_3N_4 anti-reflection coating was taken into account by an improvement in transmission of the GaAs–air interface by a factor of 1.6 [10]. The agreement between experiment and theory is not so good for the electron distribution in equation (5a). In particular the larger experimental intensity on the high energy side of the spectrum indicates that the radiative recombination rate, and with it the electron concentration close to the surface, from where the high energy photons with their small penetration depth must come, is larger than predicted by equation (5a). This conclusion is independent of the precise value of the absorption coefficient α , as long as $1/\alpha$ is much smaller than the width of the p region, which is the case on the high energy side of the spectrum. The distribution of equation (5a) must therefore be dismissed as not describing well the actual electron distribution. The agreement between experiment and theory is good for the electron distribution given by equation (5b). The small differences between calculation and measurement could be due to the use of absorption data not exactly pertinent to our diode material, as well as to the approximations to $\alpha(x)$ and $\mu_r(x)$ used in deriving equations (11).

3.2.2. Low temperatures. Figure 5 shows the results at $T = 82$ K. The theoretical calculation faces the problem of employing correct absorption data. As the diffusion length for $T = 82$ K is only $\frac{1}{3}$ of the value at $T = 296$ K [11, 12], the optically active layer of the p region is thin and close to the pn junction, where the doping concentration can only

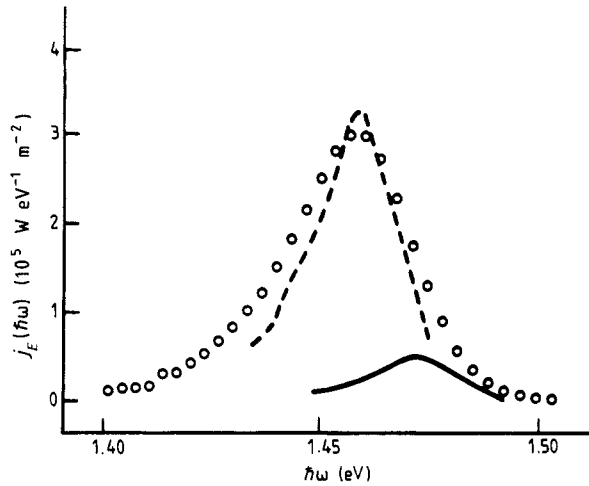


Figure 5. The experimental (circles) and theoretical (full curve) absolute spectral energy current density emitted by a LED into 2π as a function of photon energy $\hbar\omega$ at $T = 82$ K and a voltage of 1.4325 V. A better theoretical curve (broken curve) results from shifting the absorption edge to lower energy by 13 meV, corresponding to a lower Zn doping concentration at the pn junction than assumed for the full curve.

be guessed. We have used the absorption data from [13] for partly compensated p-type GaAs:(Zn, Si) for doping concentrations of $N_D = 5 \times 10^{17} \text{ cm}^{-3}$ and $N_A = 1.6 \times 10^{18} \text{ cm}^{-3}$. The agreement between experiment and theory represented by the full curve in figure 5 is unsatisfactory. The electron distributions in equations (5a) and (5b) give identical results as is expected when L_e is much smaller than the width d of the p region.

Comparing the measured and calculated curves in figure 5 it is noticed that the observed emission peak occurs at lower photon energies than calculated. It is known from [12] that a shift of the absorption edge to lower photon energy corresponds to a smaller Zn concentration than assumed above. For lack of better data we have shifted the whole energy dependence of the absorption coefficient $\alpha(\hbar\omega)$ to lower energies by 13 meV, without altering it otherwise. A recalculation of the emission spectrum results in a much better agreement with the experiment, as shown by the broken curve in figure 5. Not only is the emission peak shifted properly on the photon energy axis as intended, but also the intensity is increased by a factor of six because of its exponential dependence on the photon energy $\hbar\omega$ in the emission formula (12).

4. Conclusions

It has been shown that the light emitted by a light emitting diode is well described by a simple thermodynamic theory extending Planck's radiation law to luminescent radiation by introducing a non-zero chemical potential of photons. It predicts that the light emitted by a light emitting diode with direct band gap and given geometry is fully described by the absorption coefficient of the light generating region, the applied voltage and the temperature. These predictions have been proven experimentally for GaAs LEDs.

Regarding the uncertainties in the absorption data available in the literature, particularly at $T = 82$ K, the agreement between measured and calculated absolute values of the spectral intensity is good. It is remarkable that the presence of non-radiative transitions, usually accounted for by the quantum efficiency as a fitting parameter when relating the light intensity to the diode current, and also thought to alter the voltage dependence [14], is in fact irrelevant when relating the intensity to the diode voltage.

Acknowledgment

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