# COMMENTS AND ADDENDA

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## Distribution function of photoexcited carriers in highly excited GaAs

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We have extended our previous measurements of the radiative recombination from photoexcited hot carriers in GaAs to excitation intensities as high as $6 \times 10^6$ W/cm$^2$. We find that for $I \geq 2 \times 10^5$ W/cm$^2$, the carrier distribution is no longer Maxwellian. Another interesting result is that the emission extends well into the red region of the spectrum. Our results can be used to estimate the photoexcited carrier densities at such high intensities.

In a previous study,[1] Shah and Leite found that the electron-hole distribution function in photoexcited GaAs is essentially Maxwellian at intensities up to $\sim 5\times10^3$ W/cm$^2$. We have extended these measurements to $I=6\times10^6$ W/cm$^2$ and find that the distribution is no longer Maxwellian for $I\geq2\times10^5$ W/cm$^2$. This is somewhat surprising because one would expect that the more frequent electron-electron collisions at higher electron densities would tend to maintain a Maxwellian distribution. An interesting consequence of the non-Maxwellian carrier distribution is that the emission extends up to about 2 eV, i.e., well into the red region of the spectrum. We also observe departure from the usual logarithmic dependence of the electron temperature on excitation intensity[1] and make use of it to estimate the electron density in this region.

The experiments were performed on an $n$-type epitaxial GaAs with $\mu_e \sim 60\,000$ cm$^2$/V sec at 77 °K. The crystal was immersed in superfluid He (< 2 °K). Excitation source was a cavity-dumped argon laser, capable of giving ~50 W in 15-nsec-wide pulses at up to $10^6$ pulses per second. A double monochromator (with spectral resolution $\approx 3$ Å), an extended S-20 photomultiplier, and a gated photon-counting detection system were used to obtain the data.

Figure 1 shows the high-energy tail of the luminescence spectra at several excitation intensities up to $2\times10^5$ W/cm$^2$. We see that the luminescence intensity decreases exponentially with the increase in photon energy. If the electrons and holes are thermalized among themselves and their distribution is Maxwellian, this is precisely the shape that we expect for the high-energy emission tail.[1] We can therefore deduce that electrons and holes are indeed thermalized among themselves and can assign a temperature $T_e$ to them, following Ref. 1.

The reciprocal of these temperatures is plotted against the excitation intensity in Fig. 2. For intensities $< 10^4$ W/cm$^2$, we find that $I \propto \exp(-\hbar\omega_{LO}/kT_e)$, in agreement with the results of Ref. 1. The slope of this curve is $\approx 36$ meV, the energy of the LO phonon in GaAs. It was shown in Ref. 1 that this behavior can be explained quantitatively on the basis of polar optical scattering as the dominant energy-loss mechanism for the hot electrons. Instead of repeating these arguments here, we will examine the behavior for $I > 10^4$ W/cm$^2$.

We see from Fig. 2 that for $I > 10^4$ W/cm$^2$, the logarithmic dependence on excitation intensities does not hold. The temperature rises slower with $I$ than at lower intensities. This behavior can be caused by one of the following: (i) a more efficient loss mechanism becomes effective at these higher intensities, or (ii) one of the assumptions made in the theory of Ref. 1 is not valid at higher intensities. We will show below that the deviations in Fig. 2 can be explained by replacing an approximate expression by a more appropriate one in the

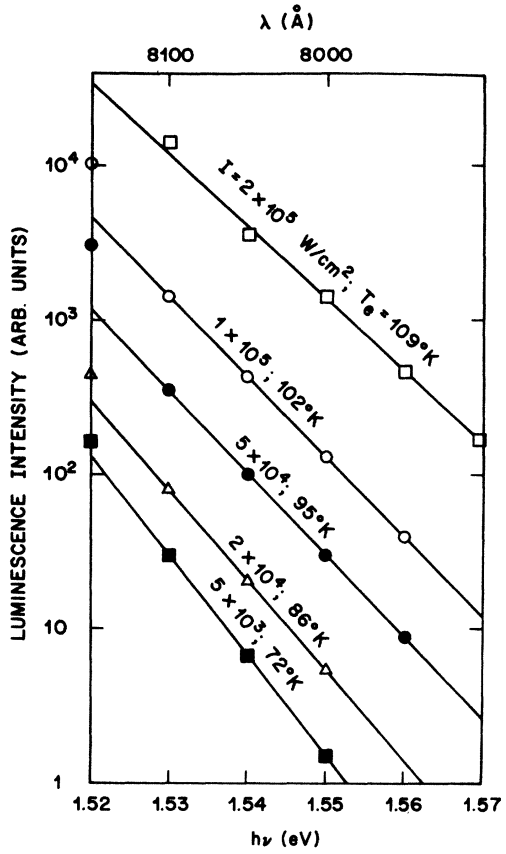


FIG. 1. High-energy luminescence tail in GaAs. The excitation intensity and the effective electron temperature are indicated on each curve.

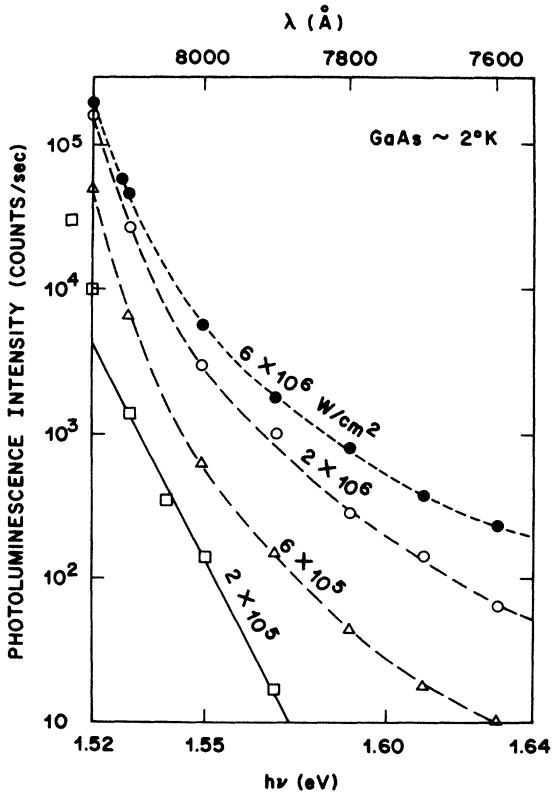


FIG. 3. High-energy luminescence tail in GaAs. Note that the luminescence intensity does not decay exponentially with photon energy for $I > 2\times10^5$ W/cm$^2$.

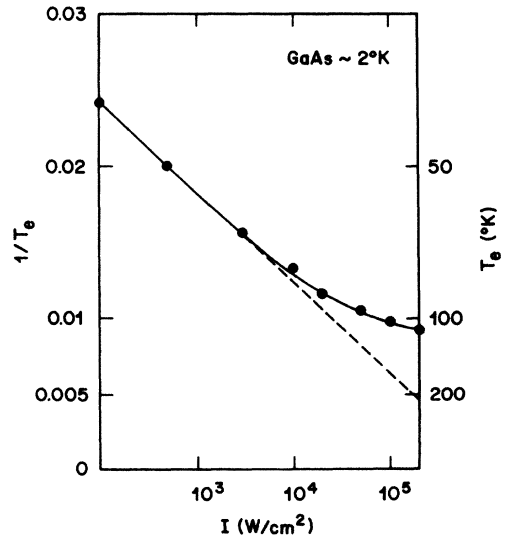


FIG. 2. Effective electron temperature vs excitation intensity $I$. The dashed curve is an extrapolation of the low-intensity data.

theory of Ref. 1.

It was shown in Ref. 1 that a photoexcited electron with excess energy $\Delta E$ gives only a fraction $[n/(n+n_c^*)]\Delta E$ to the electron gas. $n_c^*$ is $\sim 7\times10^{17}$ cm$^{-3}$ for GaAs, so the $n$ in the denominator may be neglected for $n \ll 7\times10^{17}$ cm$^{-3}$. The form $I \propto \exp(-\hbar\omega_{LO}/kT_e)$ is obtained theoretically only when $n$ is neglected in this manner. While the neglect of $n$ compared to $n_c^*$ was valid for the intensities used in Ref. 1, it may no longer be so at the higher intensities used in the present experiments and may be the cause of the observed results in Fig 2 for $I > 10^4$ W/cm$^2$. We have calculated that the observed departure from logarithmic dependence can be completely explained at $I = 2\times10^5$ W/cm$^2$ if we have $n/n_c^* = 5$ at this intensity. This would imply $n \sim 3\times10^{18}$ cm$^{-3}$, or electron lifetime in conduction band $\tau \sim 0.7\times10^{-9}$ sec. This lifetime is reasonable and gives support to the hypothesis that increasing $n$ is responsible for the observed result. The physical explanation for this is that power transferred *per electron* in the electron gas from the photoexcited electron increases linearly with intensity at low intensities, but sublinearly

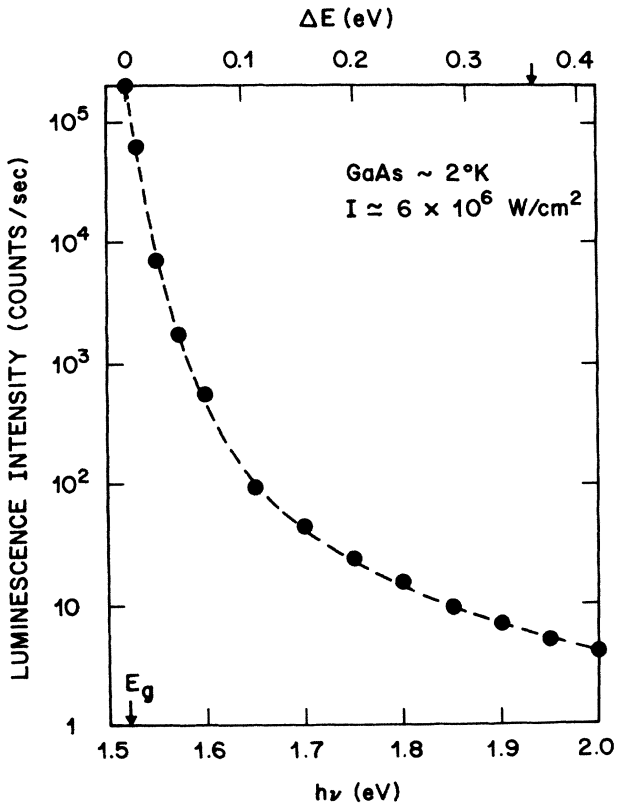


FIG. 4. High-energy luminescence tail in GaAs. The data have not been corrected for the reabsorption and the photomultiplier response. These two corrections tend to cancel each other. The scale on the top indicates the electron excess energy for a given photon energy, assuming parabolic bands and $m_e/m_h = 0.14$. The arrow indicates the energy difference between $\Gamma$ and $X$ minima of the conduction band according to Ref. 3.

with intensity at higher intensities. This results in a slower rise of $T_e$ with $I$. Similar results have been observed in CdSe[2] but at much higher intensities because $n_c^*$ (CdSe) $\sim 2\times10^{18}$ cm$^{-3}$.

We now turn our attention to intensities $>2\times10^5$ W/cm$^2$. In obtaining this data, we have chosen the repetition frequency in each case in such a manner that the *average* laser power incident on the sample is the same in each case, although the peak intensity is different. We see in Fig. 3 that while the luminescence for $I=2\times10^5$ W/cm$^2$ varies exponentially with photon energy, the other three traces show significant nonexponential behavior. Thus the electron distribution function becomes non-Maxwellian at higher excitation intensities. This result is rather surprising because one would expect that the more frequent electron-electron collisions at the higher electron densities would tend to maintain Maxwellian distribution. This points to the need for a better theoretical understanding of the photoexcited carrier distribution functions.

It should be emphasized here that the signal at higher energies ($>1.56$ eV) is *not* due to the electrons in "transit," i.e., those photoexcited electrons which are in the process of losing their large excess energy and are on their way to the bottom of the conduction band. This is because we have maintained the same average laser power in each case and yet obtained an increase in the number of photons emitted per second as the peak intensity is increased. Thus we believe that the observed emission reflects a steady-state population (within the 10-nsec pulse) of electrons and not the "transit" electrons. Since a larger fraction of the energy of the photoexcited electron is given to the electron gas at higher excitation intensities, it is not surprising that a steady-state electron population exists at higher energy. However, as we pointed out earlier, it is surprising that this distribution is non-Maxwellian.

One can see from Fig. 3 that there is a reasonably strong ($>200$ counts/sec) signal at 1.64 eV for $I=6\times10^6$ W/cm$^2$. In Fig. 4, we have plotted this curve on a compressed scale to much higher energies. We can see that the emission extends well into the red region of the spectrum. Thus some electrons exist with very high excess energy. It is interesting to note that the $X$ minimum in GaAs is $\sim 0.36$ eV above the $\Gamma$ minimum.[3] If an electron in the $\Gamma$ valley with excess energy equal to 0.36 eV were to recombine with a hole, the energy of the emitted photon (assuming parabolic bands) would be $\simeq 1.93$ eV. We have, in fact, seen emission at energies as high as this. The emission is a monotonically decreasing function of energy, indicating that the presence of the $X$ minimum at the same energy does not introduce any detectable structure in the electron occupation of those states.[4] One final point of interest is to note that there is no oscillatory structure in the high-energy emission related to the phonon energy.

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