Photoelectron energy distribution and spin polarization from activated gallium arsenide
H.-J. Drouhin, C. Hermann, M. Eminyan, G. Lampel

To cite this version:
Photoelectron energy distribution
and spin polarization from activated gallium arsenide

H.-J. Drouhin (*), C. Hermann (*), M. Eminyan (**) and G. Lampel (*)

(*) Laboratoire de Physique de la Matière Condensée (+), Ecole Polytechnique, 91128 Palaiseau, France
(**) Laboratoire de Physique de la Matière Condensée (+), Ecole Polytechnique, 91128 Palaiseau and Equipe de Physique Atomique, Tour 24, Université Paris VII, 75251 Paris 05, France

(Reçu le 25 août 1983, révisé le 25 octobre, accepté le 2 novembre 1983)

Résumé. — Des mesures de distribution en énergie à haute résolution et de polarisation de spin ont été effectuées sur des photoélectrons émis par un cristal d'arséniure de gallium activé. Les effets d'électrons non-thermalisés, l'importance de la relaxation de spin pour les électrons chauds ainsi que l'influence de la région de courbure de bande sont mis en évidence.

Abstract. — High resolution electron energy distribution and spin polarization measurements of photoelectrons emitted from an activated GaAs crystal are reported. Non-thermalized electron effects and the significance of hot electron spin relaxation as well as the influence of the band bending region are evidenced.

1. Introduction.

It has been shown [1-4] that a p-type gallium arsenide crystal activated to negative electron affinity (NEA) under circularly polarized light excitation is a convenient source of spin polarized electrons. Energy Distribution Curves (EDC) have been investigated by James and Moll [5]. Polarization of hot luminescence studies [6] proved the importance of non-thermalized electrons in the bulk crystal. Yet little work on the source itself has been published [7].

We have performed very high resolution energy analysis [8] together with spin polarization measurements of the photoelectrons emitted by a NEA GaAs crystal. We obtain very narrow EDC and relate the observed structures to the variation of the polarization with electron kinetic energy. The significance of non-thermalized electron effects and the rôle of the band bending region on the energy and spin relaxations of the photoemitted electrons are emphasized [9, 10].

2. Experiment.

In the experiment, the light is directed normal to the crystal surface. The photoemitted electrons are energy selected by a cylindrical 90° electrostatic spectrometer operating in the constant energy mode [8]. The Full Width at Half-Maximum (FWHM) of the transmission function is $\Delta E = 20$ meV, a value which has been verified experimentally by an original method using the

(+ Groupe de Recherche 050038 du CNRS.)
thermionic emission of a tungsten cathode [8]. In these conditions the transmitted current through the selector (about $10^{-3}$ of the emitted current) may exceed 1 nA without EDC distortion. EDC derivatives are obtained by applying a 10 mV peak-to-peak modulation on the photocathode biasing voltage.

Spin polarization measurements of the energy selected electrons are achieved using a Mott polarimeter with concentric cylinder electrode geometry [11, 12].

The GaAs samples, of photomultiplier type, are p-doped with zinc ($\sim 10^{19}$ cm$^{-3}$) and oriented parallel to the (100) plane. They are activated by caesium and oxygen coadsorption (at a pressure in the low $10^{-10}$ and $10^{-9}$ torr range resp.) [13]. With no caesium excess (which is our experimental condition) the total emitted current decreases with time at a rate of a few percent per hour. All the experimental curves presented in this paper are obtained under He-Ne laser excitation with energy $h\nu = 1.96$ eV, for which the quantum yields range from 1 % to 5 %. On different crystals, similar features are observed although the quantitative results are sample-dependent, as expected from the extrinsic nature of the characteristic times. More complete results will be published elsewhere.

3. High resolution energy analysis.

EDC have been measured for various excitation energies and at different temperatures. This technique allows us to distinguish between the structures related to the photon energy, i.e. to final states of optical transitions, and those appearing at a well defined energy, due to band structure properties [14].

On the EDC derivatives presented in figure 1a and figure 2 (curves $b$ and $b'$), the following typical structures are observed:

- An intense low energy peak from electrons which are thermalized in the bulk at the bottom of the conduction band: FWHM 110 meV at 300 K and 60 meV at 115 K. The FWHM is approximately the energy separation between the two extrema of the curves.

- A shoulder, whose location does not depend on the excitation energy, in the central part of the distribution. It is due to the emission of electrons initially relaxed in the L valleys of the bulk crystal. This relaxation channel is highly probable. Indeed efficient energy relaxation mechanisms for conduction electrons are collisions with optical phonons of energies close to 30 meV and the transition rate for the diffusion from $\Gamma$ to L ($\sim 1 \times 10^{13}$ s$^{-1}$) is approximately one order of magnitude larger than the inverse diffusion rate. Moreover the collision rate is $\sim 1 \times 10^{14}$ s$^{-1}$ in L compared to $\sim 1 \times 10^{13}$ s$^{-1}$ in $\Gamma$ [13]. Note that direct emission is forbidden for L valleys and the (100) face (following the theoretical treatment given in [15]). Then, these electrons are probably scattered into L prior to emission into vacuum.

- A high energy zone due to hot electrons. At 115 K, optical phonon absorption is negligible so that each collision corresponds to an energy loss. This is not the case at room temperature. As a result, a steep starting point on the EDC derivative is observed only at 115 K ($\Gamma_{8h}$ point on Fig. 1a). It corresponds to the initial kinetic energy $e_i$ of electrons created from the heavy hole band with excitation energy $h\nu : e_i = (h\nu - E_G) (1 + m_c^*/m_{hh}^*)^{-1}$ where $m_c^*$ ($m_{hh}^*$) is the conduction (heavy hole) mass and $E_G$ the forbidden gap. We verified that its energy position varies linearly as a function of $h\nu$ with the theoretical slope $(1 + m_c^*/m_{hh}^*)^{-1} \approx 0.87$ [16]. On average, the hot electrons undergo a definite number of collisions corresponding to a given energy loss before escaping into vacuum and give rise to a hot electron structure as seen on the EDC derivatives [5]. The $\Gamma_{8h}$ starting point provides an energy reference on the EDC at 115 K which is carried over to the room temperature results taking into account the variations of the bandgap and the Fermi level [8]. Thus we set the $\Gamma$ and L bulk crystal minima and observe that the structures labelled $\Gamma$ and L on the EDC are both located at an energy approximately 130 meV lower. This shift, not mentioned in [5], has also been reported for GaAs, Ga$_{1-x}$In$_x$As [17] and GaAs$_{1-x}$P$_x$ [18] photocathodes and has been attributed to energy losses during multiple
reflexions in the space charge region — possibly involving dimensional quantization [17] — because of the high reflexion coefficient of the semiconductor-vacuum interface.

From the 300 K and 115 K data, it is clear that L electrons and to a smaller extent hot electrons contribute mainly to the high energy region of the EDC. Consequently we are able to suppress the high energy emission by choosing photon energies small enough to prevent the population of the L valleys in the bulk crystal. Then for near bandgap excitation, the intense electron beam emitted by a GaAs photocathode is quasi-monochromatic. Such a source is very well suited as a source of monoenergetic electrons in collision experiments or energy loss spectroscopy. An improvement in intensity of more than three orders of magnitude over standard monochromatized electron guns can be reasonably expected [19].

With respect to time, the quantum yield of a photocathode decreases: this aging process can be attributed to either caesium desorption (mainly at room temperature where the initial yield is almost restored by adding caesium) or surface pollution (mainly at low temperature, due to cryopumping of residual gas, where warming up the cathode again restores most of the initial efficiency). In both cases a lower yield is a consequence of an increase in the electron affinity which acts as a high-pass energy filter and truncates the low energy part of the distribution. This affinity rise reduces the emitted current and affects the shape of the EDC. In the case shown on figure 1b the initial FWHM (a) first decreases (b) and then increases (c).

4. Polarization versus electron kinetic energy measurements.

The Polarization Energy Distribution Curves (PEDC) point out the drastic effect of spin relaxation as a function of kinetic energy both in the bulk and in the band bending region.

On the room temperature PEDC shown on figure 2 (circularly polarized He-Ne laser excitation, light power ~ 90 μW, quantum yield ~ 1.3 %), we observe three characteristic regions:

— The higher energy zone (between Γ_{81} and Γ_{8b}) is mainly due to electrons excited from the heavy hole valence band with an initial kinetic energy ε_i ≈ 480 meV. The theoretical initial electronic polarization is close to 50 % when averaged over the momentum distribution. The spin relaxation time at ε_i for the D'yakonov-Perel' mechanism is about 10^{-13} s [20, 21], a value of the order of the escape time. Thus, in our sample, even the electrons emitted without any significant energy loss are depolarized and we measure a maximum polarization of only 30 %.

— The depolarization of these hot electrons is enhanced with increasing escape time i.e. with increased energy loss before emission. From the steep slope observed in the decrease of the polarization it is clear that at these energies an energy loss of ~ 130 meV largely diminishes the polarization.

— The central zone shows a broad plateau due to the onset of electrons created from the light hole band with a 50 % initial mean polarization. Because of the relatively small kinetic energy of these electrons the spin relaxation mechanisms are less efficient and the spin polarization keeps a significant value. However the presence of electrons arising from the heavy hole band and having suffered a strong depolarization during the energy relaxation tends to diminish the overall polarization. Moreover at these energies there is a high probability for the electrons to partially thermalize in the L minima before being emitted through the band bending region.

— The low energy part of the distribution corresponds to the main peak of the EDC originating from thermalized electrons. These electrons are excited from the upper valence bands and also from the spin-orbit split-off band with a — 100 % initial polarization. From luminescence measurements on a similar crystal [22], we deduce a spin polarization of the electrons thermalized in the bulk close to 2 %. Using a diffusion model the emitted fraction of these electrons would have a polarization in vacuum of about 5 % [4, 7]. However, we concluded from the EDC that the electrons contributing to the Γ structure relax their energy in the band bending region. There, an electron close to the Γ minimum in the bulk crystal gets a kinetic energy of approximately 0.5 eV (the band bending) and looses about 130 meV before being emitted. The depolarization processes in the band bending region are then very similar to those discussed
previously for hot electrons created from the heavy hole band. This explains why we measure an electron polarization very close to zero.

From the PEDC, the well established increase in polarization with aging of the photocathode under light excitation of significantly more energy than the bandgap [23, 24, 9] is easily understood. It is a consequence of the crucial dependence of the emitted electron mean polarization on the electron affinity.

![Graph showing intensity derivative vs. energy above \( \Gamma_8 \) (eV)](image)

**Fig. 1a.**
Fig. 1. — EDC derivative and EDC at 115 K, with $\Delta E = 20$ meV, of electrons emitted from NEA GaAs ((100), p-type $\sim 10^{19}$ cm$^{-3}$) for 1.96 eV photon energy. Electron energy is referenced to the valence band maximum $E_g$. The experimental resolution is represented by the bracket. 

a) EDC derivative. The $I$, $L$, hot electron structures and the steep starting point $I_{ab}$ are shown (solid arrows). The theoretical positions [16] of the $I$ and $L$ extrema in the bulk crystal are marked by dashed arrows. 

b) EDC. Note the narrow low energy peak and the non-thermalized electron contribution. As the affinity rises the low energy side of the EDC is truncated. A dashed vertical line, together with the unaffected high energy side, schematizes the modified shape of the EDC. The FWHM of the initial (a) and the truncated (b and c) EDC are denoted by the horizontal bars. Cases (b) and (c) show that such an affinity cut-off can lead to a non-monotonous variation of the FWHM with aging of the photocathode.

We also used more energetic krypton-laser lines for light excitation. The PEDC show structures which can be understood in the same way as for He-Ne excitation but the hot electron polarization is reduced when the excitation energy increases. At 2.33 eV it is $\sim 25\%$ and only $\sim 18\%$ at 2.61 eV. For kinetic energies slightly higher than the excitation energy we also observe a small decrease in the polarization. At lower temperatures the electrons are created with a
Fig. 2. — EDC (a), EDC derivative (b and b') and PEDC (c) at 300 K. The initial kinetic energy of electrons excited from the heavy hole, light hole and spin-orbit split-off valence bands is respectively labelled $T_{8h}$, $T_{8l}$ and $T_{7}$. The $T$, $L$ and hot electron structures are indicated on curves b and b'.

$T = 300 \, \text{K}$
$h\nu = 1.96 \, \text{eV}$
lower kinetic energy due to the variation of the bandgap with temperature: measurements at 115 K for 1.96 eV excitation show an increase of the hot electron polarization up to 37%. All these results are consequences of the spin relaxation enhancement with increasing kinetic energy.

5. Conclusion.

EDC of NEA GaAs photocathodes measured for the first time with a very high energy resolution, and a laser excitation at different photon energies have confirmed previous results [5, 13]. However in our experimental conditions we are able to obtain much narrower electron energy distributions, leading to new monochromatic electron sources.

From the PEDC it is clear that the spin relaxation of conduction electrons at kinetic energies of a few tenths of an electron volt plays an essential rôle in determining the polarization of the photoemitted electrons, in complete agreement with theoretical predictions [20, 21]. These new studies world appear to give us powerful tool for the investigation of the photoemission properties of semiconductors and improve the characteristics of the polarized electron sources.

Acknowledgments.

We are indebted to D. M. Campbell for supplying us with his new Mott polarimeter and for useful advice. We gratefully acknowledge J.-N. Chazalviel, G. Fishman, D. Paget and C. Piaget for fruitful discussions. We thank C. Vasseur for technical help.

References


