Photoelectron scattering in a p-GaN(Cs,O) photocathode

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Abstract. Photoelectron scattering in a p-GaN(Cs,O) photocathode was studied by photoelectron emission spectroscopy at low temperatures. It has been experimentally established that the transfer of photoelectrons from the bulk of a p-GaN(Cs,O) photocathode to vacuum is accompanied by the emission of a cascade of optical phonons in the bulk and at the GaN–vacuum interface. Based on how the high-energy features of the photoelectron energy distribution shift with the photon energy, we find $m_{hh}/m_0 = 2.3 \pm 0.3$.

1. Introduction

The GaN photocathodes with effective negative electron affinity (NEA) are one of the key elements of the ultraviolet position-sensitive single-photon detectors [1] and are promising candidates for electron sources in photoinjectors [2]. Despite the practical usefulness, the influence of certain physical processes on the low-energy photoelectron escape from a GaN NEA photocathode into vacuum is still unknown. One of these processes is inelastic photoelectron scattering in the bulk of GaN and near its emitting surface.

Nonequilibrium electron dynamics in GaN was studied earlier in many theoretical and experimental works (for example, [3–5] and [6–9], respectively), but their results are partly controversial. For instance, the authors of [8, 9] conclude that the electron scattering into the side valleys in GaN is stronger than the LO-phonon scattering for electrons with energies above 1 eV, while in [10] authors state that this conclusion contradicts the calculations of the band structure and hot electron transport in GaN based on a variety of experimental findings. In [10], the authors conclude that scattering into the side valleys should start at electron energies well above 2 eV. In addition to the above-mentioned discrepancies, the calculated [3, 4] and the measured [6] phonon scattering rates also substantially differ from each other. The above examples of inconsistencies do not allow us to rely entirely on the literature data to determine the role of inelastic scattering in the process of escape of photoelectrons into vacuum from a GaN NEA photocathode. We believe that the most reliable information about this role can be obtained directly from the energy distributions of photoelectrons ($n_e(\epsilon)$) emitted from a GaN NEA photocathode to vacuum since they should contain information about the photoelectron distribution function in GaN. Earlier, $n_e(\epsilon)$ was measured in [9, 11, 12], but in these works, measurements were performed with sufficiently low energy resolution and at room temperature, which could significantly broaden the important features of energy distributions. Recently, we measured the low-temperature photoemission spectra of a p-GaN(Cs,O) photocathode with high energy resolution [13]. The measurements of the quantum efficiency ($QE(\hbar\omega)$) and the longitudinal energy distribution of photoelectrons ($n_e(\epsilon_{\text{lon}})$) were performed at 80 K,
which significantly reduced thermal broadening of the energy distributions and made it possible to detect the emission of a cascade of optical phonons by low-energy photoelectrons at the emitting surface of the \( p\)-GaN(Cs,O) photocathode.

In this work, the emission of optical phonons by hot photoelectrons in the \( p\)-GaN bulk and at the surface of a \( p\)-GaN(Cs,O) photocathode was observed and studied using photoelectron spectroscopy in a wide range of photoelectron energy.

2. Experimental details

The measurements of \( QE(h\omega) \) and \( n_e(\epsilon_{\text{ion}}) \) were performed with a homemade planar photodiode where a transmission-mode \( p\)-GaN(Cs,O) photocathode and a metallic anode were mounted parallel to each other in a hermetically sealed titanium-aluminum case. The diameters of the photocathode and anode were 18 mm, which was larger than the distance between them by a factor of about 20. The electric field near the axis of the photodiode was sufficiently uniform, and we used the photodiode as an electron spectrometer with a uniform retarding field (URF spectrometer) [14]. The photocathode was fabricated from a \( p\)-GaN/AlN heterostructure grown on a (0001) sapphire substrate by MOCVD. The thickness of the wurtzite \( p\)-GaN layer of the photocathode was 85 nm, and the concentrations of magnesium and free holes (at 300 K) in this layer were \( 1 \times 10^{19} \) and \( \sim 2 \times 10^{17} \text{cm}^{-3} \), respectively. The thickness of the \( p\)-GaN surface and the deposition of the (Cs,O) layers were described in [15].

The measurements were performed using a setup consisting of a stabilized xenon lamp, a modulator of light, a monochromator with diaphragms on the input and output slits, quartz objectives, a liquid-nitrogen optical cryostat, a digital lock-in amplifier, several DAC–ADC units, and a computer. The photoemission was studied in the temperature range of 80–300 K. The spectral resolutions of the monochromator at the measurements of \( QE(h\omega) \) and \( n_e(\epsilon_{\text{ion}}) \) were 2 and 15 meV, respectively. When measuring \( QE(h\omega) \), an accelerating voltage of 10 V was applied to the anode. To measure \( n_e(\epsilon_{\text{ion}}) \) distributions, a retarding voltage with a controlled amplitude \( U_{\text{ret}} \) and an AC voltage with a constant amplitude of 30 meV were simultaneously applied to the anode. The amplitude of the AC component of the photocurrent as a function of \( U_{\text{ac}} \) was measured by a digital lock-in amplifier and recalculated to the \( n_e(\epsilon_{\text{ion}}) \) distribution [14]. To obtain the derivative of the \( n_e(\epsilon_{\text{ion}}) \) distribution, the second harmonic of an AC component of the photocurrent was simultaneously measured by the same digital lock-in amplifier. The energy resolution of the URF spectrometer used in this work was 30 meV. The sheet resistance of a \( p\)-GaN film at 80 K was \( \sim 1\text{ MOhm/sq} \), so in order to avoid the surface charging and the corresponding \( n_e(\epsilon_{\text{ion}}) \) distortion, the photoemission currents were kept below 20 nA.

3. Results

Figure 1 shows the results of \( QE(h\omega) \) measurements at 90 K. The shape of the \( QE \) spectrum is mainly determined by the shape of the absorption spectrum of the \( p\)-GaN layer of the photocathode. One can clearly see the threshold of band-to-band transitions, below which \( QE \) decreases exponentially. The position of the band gap energy \( (\epsilon_g) \) for this photocathode is indicated by the arrow [13]. At \( h\omega > \epsilon_g \), oscillations due to the interference of light in the \( p\)-GaN/AlN heterostructure are present in the spectrum. In optical transitions in the region \( h\omega < \epsilon_g \), photoelectrons are excited both to the tails of the density of states of the conduction band of the \( p\)-GaN layer and to the band bending region (BBR) near the emitting surface due to the Franz-Keldysh effect.

We measured \( n_e(\epsilon_{\text{ion}}) \) distributions at several \( h\omega \), which are marked by enumerated triangles in figure 1. Some of the measured \( n_e(\epsilon_{\text{ion}}) \) distributions are shown in figure 2. All distributions were measured at 90 K. For all \( h\omega \), the energy distributions are broad functions in the range from the vacuum level \( (\epsilon_{\text{vac}}) \) to the energy of the conduction band minima in the bulk of \( p\)-GaN \( (\epsilon_s) \). The position of \( \epsilon_{\text{vac}} \) was determined according to [13]. It is seen that with an increase in \( h\omega \) the high-energy edge of the distributions shifts to higher energies and the number of hot photoelectrons increases substantially. Some \( n_e(\epsilon_{\text{ion}}) \) distributions contain weak oscillations in the region \( 0.6 < \epsilon_{\text{ion}} < 1.0 \text{ eV} \).
(curves 2–4). To identify the nature of these weak features in the energy distributions, the derivatives of \( n_e(\epsilon_{\text{lon}}) \) distributions were analyzed.

![Figure 1](image1.png)  ![Figure 2](image2.png)

**Figure 1.** Quantum efficiency spectrum of a \( p \)-GaN(Cs,O) photocathode.  **Figure 2.** Longitudinal energy distributions of photoelectrons emitted by a \( p \)-GaN(Cs,O) photocathode.

Figure 3 shows the derivatives of \( n_e(\epsilon_{\text{lon}}) \) multiplied by \(-1\) for the \( h\omega \) values close to the threshold of the band-to-band transitions. For convenience, the curves are shifted vertically. The derivatives of \( n_e(\epsilon_{\text{lon}}) \) at \( h\omega > h\omega_4 \) (curves 2–8) have an oscillating character. With increasing \( h\omega \), all features of the distributions shift to higher energies. It can be seen that the curves change qualitatively in the interval \( h\omega_4 - h\omega_5 \).

![Figure 3](image3.png)  ![Figure 4](image4.png)

**Figure 3.** The derivative of longitudinal energy distributions of photoelectrons emitted by a \( p \)-GaN(Cs,O) photocathode, measured at low photon energies (\( h\omega = h\omega_1 - h\omega_8 \)).  **Figure 4.** The position of a high-energy "step" vs the photon energy.

In the \( h\omega_1 < h\omega < h\omega_4 \) range (curves 2–3), the derivatives consist of a series of equidistant peaks, the distance between which is \( 95 \pm 3 \) meV. For \( h\omega_1 < h\omega < h\omega_2 \), the amplitude of the peaks increases with an increase in \( h\omega \). At \( h\omega > h\omega_4 \) (curves 2–4), the peaks begin to broaden, and the amplitude of the high-energy peak begins to decrease so that for \( h\omega > h\omega_4 \) (curves 5–8) a high-energy peak transforms into a high-energy "step".
For $h\omega > h\omega_4$ (curves 5–8), the derivatives can be divided into two regions. At $\varepsilon_{\text{lon}} \geq \varepsilon_{\text{cb}} + 0.1$ eV (marked vertically by the dashed line), the distributions have the form of weak equidistant "steps", the distance between which is $94 \pm 2$ meV. At $\varepsilon_{\text{lon}} \leq \varepsilon_{\text{cb}} + 0.1$ eV, the value of the derivative of $n_{e}(\varepsilon_{\text{lon}})$ increases sharply, and the curves consist of a series of strongly broadened peaks, similarly to the $h\omega_4 < h\omega < h\omega_5$ region. At $h\omega \geq h\omega_6$, the position of the high-energy "step" $\varepsilon_0$, indicated by the arrow in figure 3 for curve 8, shifts linearly with an increase in $h\omega$ (see figure 4), with a coefficient of $0.92 \pm 0.01$.

The derivatives of $n_{e}(\varepsilon_{\text{lon}})$ for higher $h\omega$ are shown in figure 5. One can see that with a further increase in $h\omega$, the form of the spectra does not change qualitatively. For all $h\omega$ values in the energy range $\varepsilon_{\text{lon}} \geq \varepsilon_{\text{cb}} + 0.1$ eV, the high-energy "step" shifts to higher energies and monotonically broadens with increasing $h\omega$. To describe this broadening, we subtracted the smooth component from the derivatives and took the Fourier transform in the energy region $\varepsilon_{\text{lon}} / \varepsilon_{\text{cb}} + 0.1$ eV. Figure 6 shows the dependence of the Fourier harmonic amplitude ($A_{\text{Fourier}}$) on $h\omega$. Similarly, the dependence of $A_{\text{Fourier}}$ on the temperature was measured (figure 7) for a constant value of the initial photoelectron energy in the conduction band. Also, we can note that in the energy range $1.8 < \varepsilon_{\text{lon}} < 2.1$ eV, where the minimum of the side $U$-valleys of GaN is presumably located [8, 9] (marked by "$\varepsilon_U$" in figure 5), no specific features in the energy distributions (curves 12–15) were found.

**Figure 5.** The derivative of longitudinal energy distributions of photoelectrons emitted by a $p$-GaN(Cs,O) photocathode, measured at high photon energies ($h\omega = h\omega_6$, $h\omega_5 - h\omega_4$).

**Figure 6.** The dependence of the Fourier amplitude of high-energy "steps" on the photon energy. $T = 90$ K.

**Figure 7.** The temperature dependence of the Fourier amplitude of high-energy "steps". $h\omega(T) = \varepsilon_g(T) + 0.68$ eV.
4. Discussion

We believe that the observed features of the distributions and their behavior have a clear physical interpretation. The distance between the features in the distributions is close to the energy of the LO phonons ($\hbar\Omega_{LO} = 92$ meV) in GaN, so we associate the features with the emission of optical phonons by photoelectrons. The high-energy peaks in the derivative of the $n_e(\epsilon_{ion})$ distributions in the $\hbar\omega_1 < \hbar\omega < \hbar\omega_4$ range correspond to photoelectrons that escape to vacuum without emission of optical phonons. The lower peaks correspond to the photoelectrons that emit one or more optical phonons. We should point out that oscillations in the distributions are observed at $\hbar\omega \approx \epsilon_g$ ($\hbar\omega_1 < \hbar\omega < \hbar\omega_3$), when the energy of photoelectrons in the quasineutral part of p-GaN is less than the energy of LO phonons, therefore in this case the emission of optical phonons occurs only in the BBR and on the vacuum side of the interface [13]. The amplitude of the high-energy peak at $\hbar\omega < \hbar\omega < \hbar\omega_4$ depends on the mean free path of the photoelectrons in GaN. With an increase in $\hbar\omega$, electrons gain sufficient energy to emit an LO phonon in the p-GaN bulk. In this case, the energy mean free path decreases sharply and, consequently, the amplitude of the high-energy peak decreases (figure 3, curves 3–5).

![Figure 8](attachment:image.png)

**Figure 8.** Energy diagram of a p-GaN(Cs,O) photocathode.

Figure 8 illustrates the process of photoemission in a p-GaN(Cs,O) photocathode. At low photon energies ($\hbar\omega_1 < \hbar\omega < \hbar\omega_3$), when the photoelectron energy in the quasineutral part of p-GaN is less than the energy of the LO phonon (process "1" in figure 8), electrons reach the BBR, gain kinetic energy in the near-surface electric field and escape to vacuum while emitting a cascade of optical phonons. At high photon energies ($\hbar\omega > \hbar\omega_4$), the majority of photoelectrons produced in the quasineutral part of p-GaN first emit a cascade of LO phonons (process "2" in figure 8), and then reaches the BBR with low kinetic energy. Subsequently, these electrons emit a cascade of phonons and escape to vacuum. Along with this, a small fraction of photoelectrons is generated near the surface (process "2/g397" in figure 8) at the distances of several mean free paths from the surface and escape to vacuum while emitting several phonons. In contrast to the processes "1" and "2", these photoelectrons do not have a narrow initial energy distribution because it is smeared by the near-surface electric field. Photoelectrons excited at different positions in the BBR have different energies, therefore at high $\hbar\omega$ we observe series of "steps". The position of the high-energy "step" in the derivatives of $n_i(\epsilon_{ion})$ ($\epsilon_0$ in figure 3) coincides with the initial energy of photoelectrons in the conduction band. In the model of parabolic isotropic bands, the electron energy ($\epsilon$) increases with increasing $\hbar\omega$ according to the following law:

\[
\epsilon(\hbar\omega) = (\hbar\omega - \epsilon_0)/(1 + m_e/m_h),
\]

where $m_e$ is the electron effective mass and $m_h$ is the hole effective mass. Using the value of the electron mass in GaN ($m_e/m_0 = 0.20$) [16] and the slope coefficient of the $\epsilon_0(\hbar\omega)$ dependence (figure 4), we can calculate the effective mass of heavy holes in GaN. For energies of heavy holes in the
valence band $\varepsilon_{hh} > 15$ meV, we find $m_{hh} / m_0 = (2.3 \pm 0.3)$. It is known that the valence band structure in wurtzite GaN is non-parabolic and anisotropic near the valence band edge, and that the hole effective mass increases with increasing energy [17]. In [18], an increase in the hole effective mass from $1.2 m_0$ to $1.8 m_0$ was measured in the interval of hole energies 5–11 meV by magnetoreflectance spectroscopy. We measured the value of the heavy hole mass at slightly higher hole energies and it agrees well with the above data.

With an increase in $h\omega$ and temperature, the "contrast" of all high-energy features in the derivatives of $n_e(\varepsilon_{\text{lon}})$ decreases. Apparently, this is due to the broadening of the energy distribution of photoelectrons in p-GaN. Possible physical reasons for this broadening may be related to the anisotropy of the GaN band structure, the lifetime broadening of electronic states, and acoustic phonon scattering of photoelectrons during their motion in the BBR. In order to determine the main contribution, we plan to model the generation and transport of photoelectrons in p-GaN at different electron energies and temperatures.

5. Conclusions
The photoemission spectra of a p-GaN(Cs, O) photocathode at low temperatures were measured. In energy distributions of emitted electrons, the features associated with the optical phonon cascade emission by hot photoelectrons both in the bulk and at the surface of a p-GaN(Cs,O) photocathode were observed for the first time. It is established that the emission of optical phonons is the dominant inelastic scattering channel in p-GaN(Cs,O)-photocathodes, and it significantly affects their photoemission characteristics. The value of the heavy hole mass in GaN is determined from the dependence of the position of the high-energy features of the energy distributions on $h\omega$. No features in the energy distributions related to photoelectron scattering to side valleys of GaN were observed.

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