Infrared photoluminescence spectra measurements using boxcar integrator in the active baseline subtraction mode

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Abstract. A photoluminescence (PL) measurement method using boxcar integration in the active baseline subtraction mode has been implemented. It allows for infrared PL measurements with a low duty cycle of the pump pulse, thereby minimizing the uncontrolled heating of the structure under study. The implemented method was compared to phase-sensitive detection by signal-to-noise ratio of the resulting spectra, indicating its superior performance at low duty cycles. The decrease in uncontrolled heating of the structure under study was confirmed by observing a change in the ratio of temperature-sensitive PL peaks of a test InSb/InAs/InGaAs/InAlAs/GaAs nanoheterostructure.

1. Introduction
Photoluminescence (PL) spectroscopy is one of the most widespread methods of non-destructive investigation of optical properties of semiconductors [1]. Due to the presence of thermal radiation in the infrared (IR) range, IR PL spectra measurements have their own specificity. To remove thermal background from the obtained IR PL spectra, the technique of synchronous detection is commonly used.

The technique is based on modulation of the PL excitation, and registration of PL signal at the reference frequency by lock-in amplifier (LiA). The constant thermal background generated by the components of the measuring set-up heated to room temperature is not detected. Perfect lock-in amplifier operation can be achieved when using a harmonic signal as a reference signal. The reason behind this is that the LiA recognizes only the first harmonic term of the Fourier series of input signal at the reference frequency [2].

Traditionally, a laser pump beam is chopped by using a mechanical chopper put in front of the laser. In that case, the structure under study is excited by an alternating signal, and the PL signal has a form of the square wave. As a result, the magnitude of the signal passing the lock-in amplifier is being reduced according to an expression of the Fourier series’ first component [3]. The lowest signal decreasing is reached through the use of an alternating signal with a low value of the reverse duty cycle $S$ (the ratio of the period of the signal to the pulse width) as the reference signal.

Such measurement mode with a low reverse duty cycle (i.e. a high duty cycle) leads to excessive uncontrolled heating of the investigated structure (see e.g. [4]). In this case, the real temperature of its emitting region is unknown, since the temperature sensor is located near the substrate of the sample. The exact values of the temperature of the structure emitting region are necessary for the implementation of a number of special techniques, for example, for the method of determining the internal quantum efficiency described in [5]. To reduce excessive heating of the structure, it is necessary to reduce the duration of illumination of the structure by the pump laser. It can be done by...
lowering the duty cycle of the reference pulses. Under such conditions, the lock-in amplifier would lose most of the PL signal due to Fourier series, thereby reducing the accuracy of the results.

This paper describes the method of IR PL measurement using a boxcar integrator in the active baseline subtraction mode. This method is an alternative to the method of synchronous detection in removing the parasitic thermal background from the IR PL spectra during modulation of the laser pumping by low duty cycle pulses.

2. Experimental setup

Boxcar integrator (also known as boxcar averager and gated integrator) is a tool traditionally used in time-resolved luminescence spectroscopy [6]. It is an electronic sampling instrument that integrates the signal input over a predefined period of time (gate width) and then averages over multiple integration results. Due to its time domain operation, the boxcar integrator can equally well detect signals different in form and duty cycle.

To subtract nearly constant background from the signal of interest, a boxcar integrator can be used in standard operation mode with a high-pass filter (HPF) at the input. However, there are some cases when the active baseline subtraction mode is used instead. These include scenarios when a small laser pump frequency does not allow HPF to subtract nearly constant background signal; when a signal of interest can be removed by HPF (time-resolved spectroscopy with a long lifetime); or the case when the background signal is time variable (dynamic heating of the sample with the pump pulse).

The PL measurement setup is based on a VERTEX 80 Fourier-transform infrared (FTIR) spectrometer. For maximum efficiency in the mid-IR range, the spectrometer is equipped with a KBr beamsplitter, and a liquid nitrogen cooled InSb photodetector. The structures are excited by an 809 nm laser diode. For measurements in the temperature range from 8 to 300 K, the samples were placed in a Janis CCS-150 closed cycle helium cryostat. A SR830 Lock-In Amplifier is used for synchronous detection. The metamorphic nanoheterostructure InSb/InAs/InGaAs/InAlAs/GaAs emitting at the room temperature at a wavelength of 3.3 μm [7] was chosen as a test structure for the implementation of various methods of IR PL measurements.

Boxcar integrator used in the work has two input high pass filters – ‘10 Hz’ and ‘10 kHz’. The ‘10 Hz’ filter can’t completely remove the thermal background signal from the interferogram, and therefore from the spectrum. Our laser’s pulse repetition rate is smaller than 10 kHz, therefore, while the ‘10 kHz’ filter can completely remove thermal background, it also significantly reduces the level of the signal of interest. This reduction is avoiding by using active baseline subtraction mode. The block diagram of the implemented measurement method based on a Stanford SR250 Gated Integrator and Boxcar Averager is shown schematically in figure 1.

As can be seen from the block diagram, the signal from the pulse generator is both a reference signal for the boxcar integrator, and an input signal for the electrical switch. The electrical analog switch is controlled by a transistor-transistor logical (TTL) signal, which is generated by the integrator. A logical unity of the TTL signal opens the switch, and a logical zero closes it. Since the frequency of this TTL signal is half of the reference frequency [8], the output signal of the electric switch allows the laser to be triggered “every other time”. In other words, the integrator is triggered at twice the laser modulation rate. This ensures the operation of the active baseline subtraction mode: the instrument integrates the input voltages of the signals detected by the photodetector in the presence of the PL as well as without PL (i.e. the thermal background alone), then subtracts the integration results. Finally, the subtraction results are themselves averaged, ensuring that a low frequency noise is also diminished. At the same time, the output signal-to-noise (SNR) ratio will increase in the root dependence on the number of the subtraction results averaged [9]. This number is proportional to the measurement time, and can be selected from the front panel of the integrator.
3. Results

The spectrum of the low-temperature PL ($T = 8$ K at the temperature sensor), obtained on the FTIR spectrometer without additional amplifying equipment is shown in figure 2,a. The spectrum of all IR radiation entering the interferometer was recorded, including both photoluminescence and an unwanted thermal background (see the long-wave region, figure 2,a). The synchronous detector (figure 2,b) and boxcar integrator in the active baseline subtraction mode (figure 2,c) were used to obtain the PL spectrum without thermal background. The number of sampling units averaged by the integrator was chosen in such a way that the time of the experiment with the boxcar integrator was the same as the time spent on similar measurements using the lock-in amplifier.

Figure 2 shows that the IR PL spectra of the test nanoheterostructure obtained by using both a lock-in amplifier, and a boxcar integrator are almost identical. There is no parasitic thermal background ($E < 0.34$ eV) in the spectrum obtained with the help of either amplifying equipment. This indicates the correct implementation of the technique of boxcar integration in the active baseline subtraction mode.

To compare the operation of the amplifying equipment when processing low duty cycle signals, PL spectra of the test structure were measured in a wide range of signals’ reverse duty cycles $S$ using both methods (figure 3). In this case, the comparison of the efficiency of the amplifying equipment was carried out with respect to the signal-to-noise ratio (SNR) of the obtained PL spectra by using the following equation [10]:

$$\text{SNR} = 20 \log_{10} \left( \frac{A_S}{A_N} \right),$$

where $A_S$ and $A_N$ are the root mean square amplitudes of the signal and noise, respectively.
Figure 2. Spectra of the low-temperature PL obtained: \(a\) – without amplifying equipment; \(b\) – with the lock-in amplifier; \(c\) – with the boxcar integrator in active baseline subtraction mode.

As can be seen from the figure 3, the SNR of spectra obtained with the help of the boxcar integrator almost do not depend on the reverse duty cycle, while lock-in amplifier loses most of the PL signal, and respectively SNR of the output signal, with the increase in reverse duty cycle of the signal due to Fourier series. While at a reverse duty cycle of \(S = 2\) the lock-in amplifier processes the signal 1.4 times more effectively than the boxcar, at \(S = 50\) the boxcar integrator is 6 times more effective than lock-in amplifier. Figure 3 shows that for operating with pulse signals with a reverse duty cycle \(S\) exceeding 20, it is preferable to use the boxcar integrator.

It should be added that for the case of nearly constant background better SNR of result spectrum can be achieved using boxcar integrator in standard operation mode. To complete this task, either fast laser pump pulses or additional input high pass filters can be used.

To determine the decrease in uncontrolled heating of the structure with an increase in the reverse duty cycle of the pump laser pulses, the temperature dependences of the PL of the test nanoheterostructure were additionally measured at a reverse duty cycle of \(S = 50\). At the same time, the change in the ratio of the integrated intensity of the primary (3.17 \(\mu\)m) and secondary (1.65 \(\mu\)m) PL peaks was estimated. The primary peak is relatively stable in amplitude [11] and its position changes only slightly with varying \(T\) [12], while the secondary peak decreases strongly with increasing temperature. A comparison of the experimental dependences of the ratio of the peaks’ intensities on the reverse duty cycle and on temperature has confirmed the decrease in an uncontrolled heating of the structure.
Figure 3. Experimental dependence of the SNR of the PL spectra on the reverse duty cycle $S$: 1 – for the lock-in amplifier; 2 – for the boxcar integrator in the active baseline subtraction mode.

4. Conclusion
As a result of the work, the implemented method of boxcar integration in the active baseline subtraction mode has shown itself to be a good alternative to the phase detection method. During the experimental comparison of the operation of both methods, it was determined that using the method of boxcar integration becomes preferable when a PL signal is excited with a reverse duty cycle greater than 20. It is determined that increasing the reverse duty cycle of the pump laser pulses significantly reduces the undesired heating of the structure. These results may be of interest for PL studies where precise temperature control is needed.

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References
[1] Cardona M 2010 Fundamentals of Semiconductors (Berlin: Springer) p 312
[2] Stanford Research Systems 1993 Operation and Service Manual Model SR830 DSP Lock-In Amplifier
[3] Zhuk V V and Natanson G I 1983 Trogonometricheskie ryadi Fourie i elementy teorii approksimatii (Leningrad: izd. Leningradskogo universiteta) p 21
[4] Mironova M S, Komkov O S, Firsov D D and Glinskii G F 2014 J. Phys.: Conf. Ser. 541 012085
[5] Yoo Y S, Roh T M, Na J H, Son S J and Cho Y H 2013 Appl. Phys. Lett. 102 211107
[6] Novo B M and Pessine F B T 1993 Appl. Spectrosc. 47 2044
[7] Chernov M Yu et al 2017 J. Cryst. Growth 477 97
[8] Stanford Research Systems 1987 Operation and Service Manual Model SR250 Gated Integrator & Boxcar Averager
[9] Omenetto N 1979 Analytical laser spectroscopy (New York: Wiley) pp 506–9
[10] Ziemer R E and Tranter W H 1995 Principles of Communication Systems, Modulation and Noise (New York: Wiley) p 350
[11] Komkov O S, Firsov D D, Chernov M Yu, Solov’ev V A, Sitnikova A A, Kop’ev P S and
Ivanov S V 2018 *J. Phys. D: Appl. Phys.* **51** 055106

[12] Firsov D D, Komkov O S, Solov’yev V A, Kop’ev P S and Ivanov S V 2016 *J. Phys. D: Appl. Phys.* **49** 285108