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Final Report

Time-Resolved IR Electroluminescence Spectroscopy System

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Abstract

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This report summaries the design and construction of a time-resolved photoluminescence (PL) and electroluminescence (EL) spectroscopy system consisting of a liquid He cryostat probe station, a close-cycle optical cryostat, two monochromators equipped with visible and infrared photodetectors, and a boxcar integrator and photon counter to study optical and thermal properties of optoelectronic materials and devices. Together with other existing test tools in our labs, the system offers very flexible capabilities with a temporal resolution of 1 ns over a wavelength range from 400 nm to 12 μ m. The system is also seamlessly integrated with our current Fourier Transform Infrared spectrometer for quasi-CW PL and EL measurement up to 25 μ m. The use of a temperature variable cryostat and probe station allows all the experiments to be carried out at any given temperature between 10-450 K.

This versatile system will enable many experiments, which will benefit DoD funded research, which includes two MURI programs entitled "Semiconductor Optical Upconversion Refrigeration" and "Si Based Lasers", which are both funded through AFOSR. One of the key experiments for the "Semiconductor Optical Upconversion Refrigeration" MURI program is the study of the cooling effect in light emitting diodes. The demonstration of such an effect will have tremendous impact not only on solid state refrigeration technology but also on solid state lighting and direct solar energy conversion to electricity. The basic idea of the experiment is to develop a LED sitting in the center of a hemispherical, index matched, and suspended GaAs dome. Since such a structure can theoretically provide close to unity light extraction efficiency and the material internal quantum efficiency is close to unity at low temperatures as well, the device is expected to reveal the possible net cooling by monitoring the electroluminescence peak shift under a pulsed bias as function of time.

1. Introduction

Narrow bandgap semiconductors, $E_g=1\sim12$ µm, are widely used in IR lasers and detectors for communications, sensing, imaging, and photovoltaic devices for high-efficiency electricity direct generation. The understanding of the dynamic optical, electrical and thermal properties of these materials and devices is crucial to the optimization of the device designs. One of the most straightforward approaches to study the time-dependent optical processes is to use a streak camera to record the luminescence from a sample as a function of time. Although this method is quite powerful for visible to near IR wavelength range, it has not been used for narrow gap semiconductors beyond 1 µm, where a more sophisticated optical upconversion technique using nonlinear effects has to be adopted. Recently, due to the great improvement of InGaAs photomultiplier tube (PMT), midwave IR (MWIR, 2-5 µm) and longwave IR (LWIR, 5-12 µm) semiconductor photodetectors, the temporal resolution of the measurements in these wavelength ranges is shorter than 1 ns. With such fast response photodetectors, one can readily study many physical phenomena in devices such as laser diodes and LEDs using much simpler systems.

In this program, we designed and constructed a spectroscopy system to study the timeresolved optical spectra from optoelectronic devices at variable temperatures over a broad wavelength range, 400 nm- 12 μ m. This rather unique system enhances many research projects including two, Multidisciplinary University Research Initiative (MURI) programs funded by DoD.

2. Time-resolved luminescence spectroscopy system

2.1 System design

The overall design of the proposed system is shown in Figure 1. It consists of four key parts: i) a temperature variable cryostat probe station; ii) a monochromator with multiple gratings to cover 1-12 µm and light detection devices, including a state-of-the-art InGaAs PMT

in the near IR range (900-1700 nm) and two high-speed semiconductor MWIR and LWIR photodetectors to cover 2-12 μ m; iii) two computers and software for system control, data acquisition and processing; and iv) a pulse generator and a Boxcar integrator.

A Desert cryogenic vacuum probe station is the key part of the system, which provides great flexibility to do on-wafer testing of processed devices over a broad temperature range from 10-450K. Due to the relative easy adjustment of the probe positions on the wafer in the vacuum chamber, this system also provides excellent throughput for multiple device testing in vacuum at low temperatures without the need to warm up the samples.



Figure 1: Schematic diagram of the proposed time-resolved spectroscopy system.

Based on the given specifications and the current luminescence characterization capabilities in our group, a new system for both temperature dependent (10 K-450 K) and time-resolved EL/PL spectral measurements from visible to LWIR (400 nm- 12 μ m) wavelength ranges was built. The schematic of the system optical design and the pictures of the finished test system were given in Figure 2.

The temperature dependent EL measurement will be done using this new Desert Cryogenic four-arm probe station. The configured probes for the system include: microwave coplanar probe (up to 40 GHz), low speed and DC probe, fiber based light collection probe (9 μ m core single mode and 200 μ m multi mode). All there probes can be adjusted under low

temperature with 3 dimension freedom. The positions of all probes are interchangeable. One arm was removed from the system and a custom designed optical window and numerical aperture matched Au off-axis parabolic mirror were installed to collect light coming out of edge-emitting devices. The window material is designed to be transparent up to 12 μ m and very easy to be replaced to cover any other wavelength range if needed in the future. Sample loading for the probe station is straightforward without any need for packaging and wire bonding, which is the throughput bottleneck using conventional cryostat for EL measurement.



Figure 2: (a) The schematic optical design for the system; (b) The picture of the system control instruments; (c) The picture of the system optics, including the cryostat and two spectrometers.

The signal light coming out of the probe station is collected and dispersed by reflection optics and a spectrometer. A flip mirror is used to switch light path between two visible and IR spectrometers. For the IR measurements, a 1-meter (JY Trix 550) grating spectrometer is used to provide not only high spectral resolution but also sufficient light intensity. The spectrometer is equipped with three different gratings to cover wavelength range from 950 nm to 12 µm and dual light exit ports to enable the installation of two different detectors. The three gratings can be freely switched using computer control. To ensure a large detection dynamic range for those LEDs under different biases to study the recombination processes under low injection levels, a high sensitivity state-of-the-art IR InGaAs Photomultiplier Tube (PMT) from Hamamatsu is used as one of the photodetectors. The Hamamatsu PMT head-on module has a spectral response range from 900 to 1700 nm, which is the longest wavelength for commercial PMTs and in perfect match with the InGaAs based devices under investigation for the MURI program. The MURI program necessitates the study of devices with even narrow bandgaps in the MWIR to LWIR range. Due to the lack of PMT detectors in those spectral bands, we choose to use highspeed MWIR and LWIR semiconductor photodetectors integrated with low-noise high-speed preamplifiers. In this system, three 100 MHz MCT detectors with optimized response wavelength ranges (2-5 µm, 5-8 µm, 8-12 µm) from Kolmar Technology were chosen. This is the fastest MCT detector commercially available. We worked together with the spectrometer manufacture to design the detector mount so that the changing of detector only takes less than 1 minute.

For the visible light measurement, we use an existing Spex (now part of JY) 0.34 m spectrometer equipped with a GaAs S1 PMT. A Janis close cycle cryostat with working temperature 10 K-450 K was chosen for PL setup. Right now, the PL pumping source is semiconductor laser diode because it is easy to be controlled to provide the needed light pulse width and repetition rate. Some space has been left for additional optics and a new pumping laser (see Figure 2-a).

For the electronic system, a pulse generator with short pulse width and long duty cycle is used to drive the LEDs and laser diodes. A Boxcar integrator, a photon counter or a lock-in amplifier can be freely configured to improve the signal noise ratio, to ensure the reliable detection of low-level light, and to maintain excellent temporal resolution (see Figure 2-b).

2.2 System capabilities and their potential applications

The finished system offers great flexibility for many kinds of experiments. Together with other existing test tools, the system offers the following capabilities.

- Time-resolved photoluminescence with a resolution of 1 ns for the wavelength range from 400 nm to 12 μ m. This kind of measurements can be used to study the certain carrier dynamics and thermal effects.
- Time-resolved electroluminescence measurements. This kind of measurements can provide insight into the generation-recombination processes and thermal effects in semiconductor devices such as LED and laser diodes.
- Time-resolved reflectance measurements that can reveal thermal effects in semiconductor devices, such as the cavity shift due to thermal effects in a Vertical Cavity Surface Emitting Laser (VCSEL).
- Conventional CW photoluminescence, transmission and photoreflectance measurements.
- Due to the use of a temperature variable cryostat, all the measurements mentioned above can be done over a very wide temperature range, from 10-450 K.

2.3 Custom software

Since the system is custom designed, it requires the development of computer programs to control the different parts of the system and to do data acquisition and processing. A collection of the developed software interface is given in Figure 3. The measurement programs include (a) The time resolved spectrum measurement program using a boxcar integrator; (b) The spectrum measurement program using a lock-in amplifier; (c) The time-resolved spectrum measurement program using photon counting; (d) The integrated luminescence decay time measurement using photon counting.



Figure 3. Screens of the developed measurement software; (a) main software package interface; (b) time-resolved spectrum measurement program using a boxcar integrator; (c) spectrum measurement program using a lock-in amplifier; (d) time-resolved spectrum measurement program using photon counting; (e) integrated luminescence decay time measurement using photon counting; (f) PMT noise measurement.

2.4 System function test results

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The system functions were tested using the visible-light system: a 0.34 m spectrometer, a GaAs S1 PMT, a lock-in amplifier and a photon counter. The testing results are given in Figure 4. First EL of a 650 nm laser diode (LD) was tested under low injection using a lock-in amplifier mode (Figure 4-a). The measured spectrum indicates that both spectrometer and the lock-in amplifier work normally. Then the same spectrum was measured again by changing the PMT configuration (cooling water temperature, high voltage) to photon counting mode (Figure 4-b). Next the LD spectrum at different delay time up to 100 µs and the integrated LD EL were measured (Figure 4-c and 4-d). A noise measurement program was developed to measure the counting signal level to distinguish the proper discriminator level (Figure 4-e). To further test the system functions, a time-resolved GaAs PL measurement was done. The sample was pumped by a 650 nm LD and the GaAs PL decay was measured (Figure 5). All the low temperature units (probe station and the cryostat) were also tested over the full operating range from 10K-450K. All above measurements show that all the electronics system, the control software, and cryostats work properly as designed.



Figure 4. (a) EL spectra of a 650 nm LD measured using lock-in amplifier; (b) EL spectra of a 650 nm LD measured using photon counting technique; (c) time-resolved EL spectra of a 650 nm LD measured using photon counting technique; (d) integrated long EL decay for a 650 nm LD; (e) PMT counts versus the discriminator voltage in the dark-count and signal-count modes; (f) integrated long PL decay for a 850 nm GaAs PL sample pumped by a 650 nm LD.



Figure 5. The integrated PL decay curves for GaAs and InGaAs PL samples. An overnight scan was done on B1613 GaAs PL sample to test the system stability.

3. FTIR systems

The new probe station and cryostat are seamless integrated with our current FTIR system for high-resolution spectrum measurement. A schematic drawing of the optical design is given in Figure 6-(a). For low-temperature PL measurements, the numerical aperture matched off-axis parabolic mirror P3 will collect PL light and convert it to almost parallel beam to transfer to mirror P2. Mirror P2 and P2 are aligned to con-focus and finally deliver the signal light to FTIR. Mirror P3 is mounted on a rail so that it moves freely in the horizontal direction, there will have enough space to add a room temperature PL and EL measurement system. The EL emitted from the probe station is collected by P4 and converted to parallel light and reflected by a Ag mirror M1 to P2. The system also includes one independent room-temperature white light reflectance spectrum measurement setup. Therefore, both room-temperature and temperature dependent EL and PL (10 K – 450 K) and reflectance measurement can be done easily with this setup. The switching of the configurations for the different measurements takes only a few minutes. Since the system uses all reflectance optics (gold and silver mirrors), it is easy to do high resolution (0.125 cm⁻¹) spectrum measurement for wavelength up to 25 μ m limited by the FTIR detectors and beam splitters.



A picture of the finished system is given in Figure 6-(b).

Figure 6. (a) Schematic of the integration of the probe station and the cryostat with our current FTIR; (b) photograph of the finished measurement system. The low temperature probe station is shown on the left-hand side of the picture.

4. Impact on the on-going research programs

The constructed time-resolve electroluminescence spectroscopy system has a very strong impact on several on-going research projects at ASU. In the recently awarded AFOSR MURI program, we proposed to develop semiconductor optical refrigerators based on electroluminescence upconversion (ELUC). It has been shown that a LED under a forward bias can emit photons with bandgap energy even if the bias energy is below the bandgap, namely $hv=E_g>qV_{bias}$. The basic physics of the ELUC process is schematically shown in Figure 7, where electrons (and holes) are injected and recombine in the active region. This injection process consists of three main steps: i) transport, where electrons (and holes) absorb energy from the semiconductor lattice as they progress from the metal contact into the barrier layer (TE cooling); ii) thermal relaxation, where injected electrons (holes) relax to the bottom (top) of the conduction (valence) band in the active region; and iii) recombination, where electron-hole pairs recombine either radiatively or nonradiatively. Carrier transport provides local cooling near the contacts, while relaxation and recombination generate local heating in the active region. Net cooling is only realized when the contribution of cooling is greater than the contribution of heating.



Figure 7. Schematic bandedge diagram of a semiconductor heterostructure LED under forward bias. The carrier transport and recombination processes associated with electroluminescence refrigeration are shown in the upper schematic. The spatial variation of electron (hole) average transport energies E_{Tn} (E_{T_p}) and electron (hole) average recombination energy E_{Rn} (E_{Rp}) under the isothermal conditions are shown in the lower schematic.

A proposed device structure used to demonstrate the net cooling effect is given in Figure 8. In this design, the cooling device sits on a hemispherical lens for the maximized light extraction and the whole structure is supported by four thin and narrow semiconductor beams for the minimized thermal conduction. Metal contacts are fed to the cooling device by going on the top of two beams. The finished device should be tested in vacuum surrounded by highly light absorbing materials to minimize thermal convection and radiation. In order to maximize light extraction efficiency, the device active region size and the radius of the hemispherical lens should be chosen carefully. In principle, beams with smaller cross section give better thermal isolation. On the other side, the beam cross section should be sufficiently large to provide enough mechanic strength to support the whole device structure.



Figure 8. Optical refrigeration device with suspended lens structure.

Time resolved FEA is used to model this device. The device structure plotted in Figure 8 does not have rotation symmetry and can not be calculated in a simplified 2D model. In this calculation we choose a thin disk to represent the four beams by keep the same cross section areas. The simulated device has an active region with 50 μ m radius and 1 μ m thickness. The hemispherical lens radius is 100 μ m. The equivalent cross section area is 15×10 μ m² for each beam. The cooling power density and material parameters are chosen based on our previous theoretical model. The calculated temperature profile for the device in steady state and the temperature ch^aange versus time are plotted in Figure 9-a. The calculated maximum temperature drop is 6 °C, which is measurable using our current experimental setup. Figure 9-b indicates that the temperature drop starts 10 μ s after the bias is applied and the device temperature is stabilized after 0.1 s. A significant temperature drop happens after 1 ms. The finial cooling temperature is determined by the boundary condition while the cooling time is determined by the thermal capacitance of the device.

Since the active region temperature drop can not be directly measured, the EL peak wavelength is measured to give an indirect measurement. Although the heating due to optical loss outside of the active region may prevent the demonstration of net cooling, it is still possible to see the local cooling effect within a time window of 10^{-5} s to 10^{-2} s based on the time-resolved

FEA modeling. A more detailed drawing to explain the measurement concepts is given in Figure 10. In Figure 10-a, the bottom part is the applied bias waveform and the top part is three possible temperature evolution profiles: net heating, local cooling and net cooling, from top to bottom, respectively. Figure 10-b to 10-d give the schematic drawing of the EL spectrum evolution versus time for corresponding net heating, local cooling, and net cooling cases. Using the local cooling case as an example, when bias is applied, the active region temperature will decrease and the spectrum will show blue shift first. When the heat generated outside of the active region due to optical absorption is conducted back to the active region, the temperature will increase and the spectrum will show red shift. Therefore, time-resolved spectra measurement is a powerful method to investigate the local cooling effect.



Figure 9. Time resolved FEA for the suspended lens cooling device; (a) temperature profile when device is in a stead state; (b) temperature drop versus time.

Another important factor to demonstrate net cooling is the material internal quantum efficiency. Using current low temperature, we have estimated the material internal quantum efficiency versus temperature, which is given in Figure 11. The experiment shows that the internal quantum efficiency tends to approach to unity when the temperature drops down to 100 K. This measurement gives the working temperature window to observe net cooling. When temperature is high, the internal quantum efficiency is low and there is no net cooling; when temperature is low, even net cooling can easily happen; the cooling power is low due to a low cooling efficiency. Therefore, measuring the time resolved EL spectrum at temperature range 50-100 K will give a great chance to first see the net cooling.



Figure 10. Expected time-resolved spectral measurements for local cooling.



Figure 11. Temperature dependent internal quantum efficiency of a GaAs PL sample versus pumping laser power.

5. Conclusion

A time-resolved PL and EL system has been built to study optical and thermal properties of optoelectronic devices. The system offers very flexible capabilities for time-resolved PL and EL measurements with a temporal resolution of 1 ns over a wavelength range from 400 nm to 12 μ m. The system is also seamless integrated with our existing Fourier transform infrared spectrometer for quasi-CW PL and EL measurements up to 25 μ m. The use of a temperature variable cryostat and probe station allows all experiments to be carried out at any given temperature between 10-450K.

This versatile system enables many experiments, which will benefit DoD funded research, including two MURI programs entitled "Semiconductor Optical Upconversion Refrigeration" and Si Based Lasers", which are funded through AFOSR. An immediate application of this unique system is to use low temperature time-resolved EL spectroscopy to study electroluminescence refrigeration in LEDs.

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