The complex physics of photoinduced charge carriers in novel materials

Summary

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Semiconductors are the basic building blocks of our modern technology. Semiconductors are used, among other applications, in thin-film transistors, light-emitting diodes, and solar cells. The industry and scientific community constantly seek new materials and fabrication methods that result in more efficient, reliable, and cost-effective devices. Observing photoinduced change in semiconductors as a function of temperature may lead to a deeper understanding of the processes responsible for the excitation and recombination of charge carriers. By understanding the physical processes responsible for the observed recombination dynamics, research can focus on materials with the desired properties. The characterization and comparison of wide-bandgap semiconductors can be difficult due to the persistent photoconductivity present in metal oxide semiconductors. The reliable characterization and comparison of samples with persistent photoconductivity are important for the development of devices based on these materials and for the understanding of the physical processes governing the prolonged excitation and relaxation process. In my thesis, I present the measurement of temperature-dependent charge-carrier lifetime in metal halide perovskites and the observation of the persistent photoconductivity effect in metal oxide semiconductors.

I developed two measurement systems for the detection of temperature-dependent charge-carrier recombination dynamics. Both measurements are based on detecting changes in the microwave signal reflected by the sample under investigation. The change in reflected microwave signal is proportional to the change in the charge carrier density, thus it is possible to observe the recombination process through the measurement of the microwave signal. One of the systems is based on a coplanar waveguide, and the other is based on a microwave cavity with a high Q-factor. The cavity-based instrument was built with the high Q-factor microwave cavity and microwave bridge with automatic frequency control of a commercially available electron spin resonance spectrometer. The cavity allows for the quick cooling of the sample. The coplanar waveguide-based system is able to measure samples in any geometry with the cooling being done through a cold finger. The frequency of the probing electromagnetic radiation can be tuned in a wide range thus determining the penetration depth into the material. The coplanar waveguide-based system is equipped with a reference arm that cancels out the reflected microwave signal occurring without illumination, preventing measurement saturation and increasing the signal-to-noise ratio.

In my thesis, I present the temperature-dependent recombination dynamics of photoinduced charge carriers in metal halide perovskites in the methylammonium lead halide group and cesium lead bromide. I observed ultralong charge carrier recombination times in methylammonium lead halides single crystals in the orthorhombic phase with the longest recombination time being over 68 μ s in the MAPbBr₃ perovskite. I further investigated the effect of disorder in the methylammonium lead halides by carrying out temperature-dependent recombination time measurements on samples with differing morphology and comparing recombination time and photoconductivity in a sample after slow cooling and quenching the crystal structure. From these measurements, I found that the increase of grain boundaries lowers the recombination time. I observed ultralong charge carrier recombination times in single crystal samples of the CsPbBr₃ perovskite. I ruled out the effect of sample heating, compared the results with transient photoluminescence measurement, and conducted steady-state photoconductivity measurements to support the results. I analyzed the temperature-dependent and excitation power-dependent charge carrier recombination time is caused by the detrapping of charge carriers limiting the recombination process.

I developed a measurement protocol for the comparison of samples exhibiting the persistent photoconductivity phenomena. I compared a series of Indium Gallium Zinc Oxide thin film samples using an illumination-relaxation cycle to demonstrate the method. I demonstrated the reproducibility of the measurements and compared the observed tendencies in the sample series with the information found in the literature.