Summary

This work represents a collection of scientific results that were obtained by me together with my research collaborators in the last 7.5 years. This period I have been a postdoctoral fellow in the USA and Germany and a research assistant in Romania. These results have been published during last years in various international journals of good scientific impact factor. Results have been combined together to form few chapters and sub-chapters which in turn would give us a good chronological understanding of how is the precise control of chain conformations, of various conjugated macromolecules, impacting the final optoelectronic properties of such materials. As we will see, this precise control of molecular conformations can be realized using a variety of polymer ordering and processing methods. This includes the few popular methods such as the exposure of the material to solvent vapor, phase separation, induced crystallization, dewetting or even novel methods such as the recently reported method of exposing thin films of conjugated polymers to light.

Therefore, a first part of this thesis contains results that have been obtained by using a rational, innovative design to develop hybrid nanostructures (consisting of conjugated donor polymers and acceptor materials such as fullerenes) as well as organic nanostructures (entirely made of conjugated donor-acceptor diblock copolymers) that can be used in nanotechnological applications such as hybrid or organic solar energy devices.

The next part of my habilitation thesis contains results that are dedicated to the development of novel model systems of conjugated polymers by combining the crystallization method with the so-called “self-seeding” technique. The aim of this work was to obtain polymeric systems where all conjugated macromolecular chains are packed in a perfect registry at nanoscale, microscale and macroscale and thus, form defect-free superstructures containing no amorphous, bended, entangled or structurally deteriorated chains. The optoelectronic properties

(including absorption and emission) of such model systems as well as their charge transport and mobility were expected to be much improved.

The last part of this thesis represents a collection of rather recent results that were obtained using both dewetting and processing by light methods, the latter consisting in exposure of thin films of conjugated polymers to light. We will see that ordering and manipulating the structure of conjugated polymers is offering tremendous advantages in altering and tuning the final optoelectronic properties of such materials. We show that manipulation of molecular conformation of conjugated polymer macromolecules in thin films is possible not only at molecular dimension and microscale but also at macroscale and on *large-surface area*. Moreover, due to well-known structure-optoelectronic properties relationship that is valid for conjugated polymer systems, a significant improvement of optoelectronic properties is thus expected after processing. Indeed, materials that have been processed using these methods exhibited puzzling emission properties with their emission being enhanced by *50 folds*. This is emphasizing the huge potential of conjugated polymers in case they would eventually be implemented into energy devices such as light-emitting diodes or other display nanotechnology devices.

At the end of this thesis, I have included a chapter that exposes my future research directions. Here, I included the current ongoing research directions, many of them described in this work, as well as new research directions that I plan to develop in the next years. One of these new research direction aims to adapt a method that is already used to order nanoparticles of colloidal solutions on solid substrates (the so-called "convective self-assembly" method) and to use it to create new microstructures of conjugated polymers. The advantage of such a method is the possibility to precisely control specific physical parameters such as the temperature of the substrate and the deposition speed (i.e. solvent evaporation rate) and to obtain and easily reproduce ordered/oriented microstructures over an area of many squared centimeters. Another direction aims to combine conjugated polymers with metallic nanostructures based on gold

and silver nanoparticles ordered on solid substrates. The goal here is to create hybrid materials that could exhibit enhanced emission properties due to the so-called “metal enhanced fluorescence” mechanism. Also here, I have included my teaching philosophy that I use in my current teaching and that I intend to further develop in the future when I will eventually have the opportunity to teach further courses related to conjugated polymers, to their processing and properties as well as to their use in current and future technological applications.