Abstract

Two-dimensional inorganic-organic (2D-IO) hybrids are very interesting material systems, which, because of their unique stacking layered structure and tunable physical properties, have become very popular amongst application scientists and material engineers, recently. The 2D-IO hybrids exhibit tunable optical, structural, magnetic, and thermal properties which can be controlled up to a large extent in each attribute of theirs by appropriate engineering in their inorganic networks and the interplanar organic moiety during synthesis. The length and conformation of the organic molecules largely decide the overall structural and thermal stability while the type of the divalent metal and halogen ions in the inorganic network determine the optoelectronic and magnetic properties. Numerous reports in the recent literature, from the perspective of potential applications in varieties of areas such as data storage, memory devices, low-temperature magnetic refrigeration technology, energy storage and optoelectronics, show the significance of these material systems and the extensiveness for further exploration they bring in.

There are two facets of the work that has been conducted in the current thesis. The first one deals with the chemical synthesis of the new and lead-free 2D-IO hybrid systems and in the second one, investigation of the optical, magnetic and thermal properties of those have been carried out. The lead-free and transition metal-based 2D-IO hybrids have been synthesized by two-step solution processing method. Thermal stability and solid-solid structural phase transitions have been studied in the copper- and manganese-based 2D-IO hybrids by varying the length and conformation of the organic part, and metal-halogen network. The longer carbon atom chain and manganese chloride-based 2D-IO hybrids are found to be most thermally stable among all the systems studied here.

Diversity in the structural and optical properties has been investigated in the copper-based 2D-IO hybrids by varying the size of the organic part and the halogen element. The longer organic-based IO hybrids are found to be in uniform single crystalline phase whereas the shorter organic based-IO hybrids are seen to exist with composition dependent structural frustration and immiscible amorphous phase. The optical properties are independent of the organic moiety present in the hybrids. Also, the absorption features such as optical band gap can be systematically tuned by proper composition of the halogen ions. Much importantly, a broadband white light emission in the entire visible range extending into the infrared is observed from copper and manganese

based 2D-IO hybrids. The copper-based 2D-IO hybrids are ferromagnetic below the Curie temperature in the range of 7-12 K due to the Jahn-Teller distortion of copper ions whereas manganese-based IO systems are antiferromagnetic below Neel temperature of ~43 K. In the mixed Cu-Mn 2D-IO hybrid systems, intertwined magnetic phases corresponding to ferromagnetic and antiferromagnetic phases are observed at a particular Cu-Mn doping level. These results indicate that only after a certain Mn-doping level, it does not lead to Cu substitution rather prefers to form its own crystalline phase. Therefore, a competition between the structural and magnetic entropies is at the interplay. Lastly, the Magnetocaloric effect has been studied in the Cu-based 2D-IO hybrids to find their suitability in low-temperature magnetic refrigeration technology. The Magnetocaloric performance of these systems is found to be comparable to those of the more commonly studied transition metal oxide perovskites.