Modulating the emission of CsPbBr$_3$ perovskite nanocrystals via thermally varying magnetic field of La$_{0.67}$Sr$_{0.33}$Mn$_{0.9}$(Ni/Co)$_{0.1}$O$_3$

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ABSTRACT
The role of the magnetic field in the emission properties of CsPbBr$_3$ perovskite nanocrystals is investigated using magnetic materials, La$_{0.67}$Sr$_{0.33}$Mn$_{0.9}$Co$_{0.1}$O$_3$ and La$_{0.67}$Sr$_{0.33}$Mn$_{0.9}$Ni$_{0.1}$O$_3$. The ferromagnetic–paramagnetic phase transition point of these magnetic materials is near room temperature, and the intensity of the magnetic field can be controlled by changing the temperature. An increase of 51% and 33% is observed in the emission intensity of the CsPbBr$_3$ perovskites, on increasing the temperature from 10°C to 35°C, in the presence of La$_{0.67}$Sr$_{0.33}$Mn$_{0.9}$Ni$_{0.1}$O$_3$ and La$_{0.67}$Sr$_{0.33}$Mn$_{0.9}$Co$_{0.1}$O$_3$, respectively. At lower temperatures, the samples are magnetic due to their ferromagnetic nature, and on increasing the temperature, they become non-magnetic. Magnetic materials as well as CsPbBr$_3$ nanocrystals possess perovskite crystal structure, and this might be playing an important role in transmitting the magnetic field. By understanding the role of the magnetic field in the emission of CsPbBr$_3$ perovskite nanocrystals, magnetic materials can be used to control the properties of CsPbBr$_3$ nanocrystals for light energy harvesting and opto-electronic applications.

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INTRODUCTION
CsPbBr$_3$ perovskite nanocrystals have been recognized as a promising material for the fabrication of light harvesting and display devices, owing to their fascinating emission properties.$^{1-6}$ The unique features of CsPbBr$_3$ perovskites include (i) broad and size dependent absorption, (ii) high extinction coefficient, (iii) narrow emission, and (iv) high photoluminescence and electroluminescence yield.$^{7-15}$ So far, lead halide perovskites are one of the known materials that displayed the highest efficiency for photovoltaics and light emitting diodes (LEDs).$^{17-20}$ Another advantage of the CsPbBr$_3$ perovskites is the bandgap tunability by the halogen exchange reactions. The exchange of bromine ions with chlorine results in the blue shift in absorption and emission, and at the same time, the exchange with iodide ions results in the red shifted spectrum.$^{21-23}$ By the controlled exchange of halogen ions, the emission in the whole visible region can be obtained from the cesium halide perovskites, and thus, it is considered a suitable material for light harvesting and opto-electronic applications.$^{11,26-28,30,31}$

Controlling the emission intensity of the CsPbBr$_3$ perovskite nanocrystals is essential for various applications in light harvesting/display devices.$^{24}$ The efficiency of a solar cell is known to increase in the presence of an applied magnetic field.$^{33}$ The influence of the magnetic field on the CsPbBr$_3$ perovskite nanocrystals can be monitored using emission studies.$^{34}$ Even though the magnetic field can control the properties of CsPbBr$_3$ perovskite nanomaterials, it is difficult to vary the magnetic field of a single magnet. A magnetic material, with a controllable magnetic field, can be the perfect material for controlling the physical and chemical processes.$^{35}$ Thus, we have used Ni/Co doped La$_{0.67}$Sr$_{0.33}$MnO$_3$ for controlling the emission properties of CsPbBr$_3$ perovskites. The magnetic field from the Ni/Co doped La$_{0.67}$Sr$_{0.33}$MnO$_3$ can be controlled with temperature as it exhibits a magnetic caloric effect.$^{36,37}$ Upon increasing the temperature, ferromagnetic Ni/Co doped La$_{0.67}$Sr$_{0.33}$MnO$_3$...
becomes paramagnetic.\textsuperscript{36,37} The Curie temperature, ferromagnetic-paramagnetic transition point, of Ni/Co doped La\textsubscript{0.67}Sr\textsubscript{0.33}MnO\textsubscript{3} is near room temperature (vide infra). Below the Curie point, Ni/Co doped La\textsubscript{0.67}Sr\textsubscript{0.33}MnO\textsubscript{3} exhibits a permanent magnetic behavior as a result of the ferromagnetic character.\textsuperscript{36,37} Above the Curie point, this material loses its permanent magnetic behavior by converting to paramagnetic. As the Curie temperature of Ni/Co doped La\textsubscript{0.67}Sr\textsubscript{0.33}MnO\textsubscript{3} is near room temperature, this is a suitable material for modulating the emission of CsPbBr\textsubscript{3} perovskite nanocrystals.\textsuperscript{36,37} Magnetic materials showing a Curie temperature above 45 °C are not good to control the emission of CsPbBr\textsubscript{3} perovskite nanocrystals as it starts decomposition above 45 °C. Crystal structures of Ni/Co doped La\textsubscript{0.67}Sr\textsubscript{0.33}MnO\textsubscript{3} and the CsPbBr\textsubscript{3} nanocrystals are perovskite structures, and this can impart the magnetic field more effectively.\textsuperscript{36,37} By keeping all these things in mind, herein we investigate the role of the magnetic caloric effect of Ni/Co doped La\textsubscript{0.67}Sr\textsubscript{0.33}MnO\textsubscript{3} in the emission properties of CsPbBr\textsubscript{3} perovskite nanocrystals.

**RESULTS AND DISCUSSIONS**

In order to achieve the above objective, CsPbBr\textsubscript{3} perovskite nanocrystals capped with oleylamine and oleic acid were synthesized by following a reported procedure with slight modifications.\textsuperscript{40–42} Details of the synthesis and purifications are given in the supplementary material. Furthermore, the microscopic and spectroscopic characterization of these nanocrystals was carried out and presented in Fig. 1. The transmission electron microscopic (TEM) analysis clearly depicts that the nanocrystals formed are mono-disperse and crystalline in nature [Fig. 1(a)]. The average size of the perovskite nanocrystals was estimated from the histogram of the size distribution and estimated as 4.0 nm (supplementary material). Furthermore, from the high-resolution TEM analysis (HR-TEM), the d-spacing value corresponding to the (200) plane of cubic CsPbBr\textsubscript{3} was estimated as 0.58 nm.\textsuperscript{46} d-spacing analysis was carried out by following the reported literature.\textsuperscript{46} Details of the analysis are given in Fig. 1(b) and in the supplementary material. The elements present in the samples were analyzed using energy dispersive spectroscopy (EDS) and presented in the supplementary material.

Furthermore, the spectroscopic analysis showed all the characteristic features of the CsPbBr\textsubscript{3} nanocrystals. The absorption spectrum showed the presence of multiple excitonic peaks as a result of the quantum confinement effect.\textsuperscript{41,42} The first excitonic peak, the minimum energy transition corresponding to the bandgap, was centered at 495 nm [Fig. 1(c)]. Furthermore, the emission of the nanocrystals was recorded by exciting at 350 nm and keeping the excitation and emission slit widths as 2 nm. The emission spectrum of the CsPbBr\textsubscript{3} nanocrystals was found to be narrow with the maximum at 505 nm. The symmetrical emission spectrum with the full width at half maximum of around ~17 nm indicates the mono-disperity of the samples.\textsuperscript{7} The emission quantum yield of the CsPbBr\textsubscript{3} perovskite nanocrystals was estimated by a relative method using fluorescein dye as the standard. The emission quantum yield of the present nanocrystals was found to be 0.68, and the details of the quantum yield measurements are given in the supplementary material. Furthermore, Powder X-ray Diffraction (PXRD) was used to characterize the crystal structure of CsPbBr\textsubscript{3} nanocrystals. X-ray diffraction patterns clearly match with the Joint Committee on Powder Diffraction Standards (JCPDS) peak positions of the cubic CsPbBr\textsubscript{3} cubic perovskite structure and earlier reported values in the literature.\textsuperscript{47} The x-ray diffraction (XRD) peaks were observed at 2θ = 15.4° and 31.08° corresponding to (100) and (200) of cubic CsPbBr\textsubscript{3} perovskite nanocrystals [Fig. 1(d)]. Broadening in the x-ray diffraction can be attributed to the fine nanocrystalline nature.\textsuperscript{46,47}

![FIG. 1. Spectroscopic and microscopic characterizations of CsPbBr\textsubscript{3} perovskite nanocrystals. (a) Low resolution and (b) high resolution TEM images of perovskite nanocrystals. The d spacing analyses of nanocrystals are carried out along the z-axis using Gatan DigitalMicrograph software. (c) Absorption (a, black trace) and emission (b, blue trace) of CsPbBr\textsubscript{3} perovskite nanocrystals. (d) X-ray diffraction pattern of cubic CsPbBr\textsubscript{3} perovskite nanocrystals. Note: The emission spectrum of the CsPbBr\textsubscript{3} perovskite nanocrystals in chloroform was recorded by exciting the samples at 350 nm and keeping excitation and emission slit widths as 2 nm.](image-url)
well and subjected to x-ray diffraction analysis, and the results are presented in Figs. 2(a) and 2(b). The phase purity and crystal structure of the two compositions were confirmed from the Rietveld refinement of x-ray diffraction patterns using the GSAS software. The analysis of the XRD data confirmed that two compositions are single phase with perovskite structure ($R3c$ space group). The refinement was carried out using the structural parameters of $La_{0.67}Sr_{0.33}MnO_3$ as starting values. The observed, calculated, and difference pattern for the two compositions are shown in Fig. 2, and their refined structural parameters are given in the supplementary material.

Furthermore, magnetization measurements of the samples were made as a function of temperature and applied field using a vibrating sample magnetometer attached to a physical property measurement system. The temperature dependence of zero field cooled (ZFC) and field cooled (FC) magnetization of $La_{0.67}Sr_{0.33}MnO_3$ and $La_{0.67}Sr_{0.33}Mn_{0.9}Co_{0.1}O_3$ under a magnetic field of 0.5 kOe is shown in Figs. 3(b) and 3(a), respectively. Both the compounds show a near room temperature ferromagnetic–paramagnetic phase transition (Curie temperature) around $\sim 305$ K for $La_{0.67}Sr_{0.33}Mn_{0.9}Co_{0.1}O_3$ and $La_{0.67}Sr_{0.33}Mn_{0.9}Ni_{0.1}O_3$.

In order to study the influence of the thermally varying magnetic field of Ni/Co doped $La_{0.67}Sr_{0.33}MnO_3$ on the emission properties of perovskite nanocrystals, the samples were spin coated on a glass slide and the experiments were performed. If there is any interaction between the magnetic field of the magnetic material and the exciton of the semiconductor material, there will be some changes in the emission intensity. Initially, Ni/Co doped $La_{0.67}Sr_{0.33}MnO_3$ was spin coated on a glass slide, and after drying, a thin film of CsPbBr$_3$ samples was coated onto it. At lower temperatures, we observed the decomposition of the samples by the condensation of atmospheric moisture and it is already known from the literature. In order to prevent this, samples were well covered with a cover slip and the sides were covered by cellulose-based adhesive tape. Furthermore, we have measured the emission intensity variations of the samples with temperature (from 10 $^\circ$C to 45 $^\circ$C). After 45 $^\circ$C, we observed the decomposition of the perovskite samples, which is already known from the

**FIG. 2.** Rietveld refined XRD pattern of $La_{0.67}Sr_{0.33}Mn_{0.9}Co_{0.1}O_3$ (a) and $La_{0.67}Sr_{0.33}Mn_{0.9}Ni_{0.1}O_3$ (b). The experiment was carried out using a PANalytical XPert Pro diffractometer in the Bragg-Brentano geometry with Cu-Kα radiation with a step size of $2\theta = 0.017^\circ$.

**FIG. 3.** Temperature dependence of magnetization (ZFC and FC) of (a) $La_{0.67}Sr_{0.33}Mn_{0.9}Co_{0.1}O_3$ and (b) $La_{0.67}Sr_{0.33}Mn_{0.9}Ni_{0.1}O_3$ under the external magnetic field of 0.5 kOe. Both the compounds exhibited the ferromagnetic–paramagnetic phase transition around 305 K ($\sim 32$ $^\circ$C).
Interestingly, we observed two distinct types of emission variation for both the samples: an increase in emission intensity up to 35°C and, after this temperature, a small decrease in the emission intensity. Temperature induced emission changes in CsPbBr₃ perovskite nanocrystals on Ni doped La₀.₆₇Sr₀.₃₃MnO₃ are presented in Figs. 4(a) and 4(e) and that of Co is presented in Figs. 4(b) and 4(f). In order to get a better understanding of the emission properties in the presence of Ni/Co doped La₀.₆₇Sr₀.₃₃MnO₃, emission lifetimes were taken and presented in Figs. 4(c) and 4(d). As a control experiment, we have performed the temperature dependence of emission of CsPbBr₃ perovskite nanocrystals and presented in Fig. S2 of the supplementary material. If there is any interaction between the semiconductor and the magnetic material, there will be some change in the emission lifetimes. Interestingly, lifetimes of CsPbBr₃ samples showed an increased magnitude with temperature. The emission decay traces at two distinct temperatures for Ni doped La₀.₆₇Sr₀.₃₃MnO₃ showed a tri-exponential nature with an average lifetime of 8.4 ns at 10°C and 11 ns at 35°C. The average lifetime exhibited by Co doped La₀.₆₇Sr₀.₃₃MnO₃ was 8.9 ns at 10°C and 10.8 ns at 35°C. The decrease in the emission intensity and lifetime of CsPbBr₃ nanocrystals, in the presence of magnetic field, can be attributed to the formation of less emissive triplet excitons. The details of the experiments are given in the supplementary material.

The increased emission of CsPbBr₃ can be attributed to the decrease in the magnetic field and the small decrease after 35°C is due to temperature induced changes. Ni/Co doped La₀.₆₇Sr₀.₃₃MnO₃ exhibits ferromagnetic behavior at lower temperatures, and at around 32°C, it becomes paramagnetic (vide supra). In an earlier report, Kovalenko and Sá observed a decrease in the emission intensity of CsPbBr₃ perovskite nanocrystals in the presence of the magnetic field. They have observed 10% decrease in the emission yield for CsPbBr₃ nanocrystals in the presence of 500 mT. In our case, upon removal of the magnetic field, by increasing the temperature, the emission yield of CsPbBr₃ perovskite nanocrystals is found to increase by 51% in the case of Ni doped La₀.₆₇Sr₀.₃₃MnO₃ and 33% in the presence of Co doped La₀.₆₇Sr₀.₃₃MnO₃. At lower temperatures, La₀.₆₇Sr₀.₃₃Mn₀.₉Co₀.₁O₃ has an antiferromagnetic behavior, and thus, the magnetic field produced by the Co doped La₀.₆₇Sr₀.₃₃MnO₃ is lesser than that of Ni doped La₀.₆₇Sr₀.₃₃MnO₃. The crystal structure of Ni/Co doped La₀.₆₇Sr₀.₃₃MnO₃ and the CsPbBr₃ nanocrystals is perovskite structure, and this might be playing a crucial role in transmitting the magnetic field from Ni/Co doped La₀.₆₇Sr₀.₃₃MnO₃ to CsPbBr₃.

FIG. 4. Emission [(a) and (b)] and lifetime [(c) and (d)] changes in CsPbBr₃ perovskites on Ni [(a) and (c)] doped and Co [(b) and (d)] doped La₀.₆₇Sr₀.₃₃MnO₃ at 10°C and 35°C. Change in the emission intensity of the CsPbBr₃ perovskite on Ni (e) doped and Co (f) doped La₀.₆₇Sr₀.₃₃MnO₃ from 10°C to 45°C by exciting the samples at 350 nm and keeping the excitation and emission slit widths as 3 nm.
CONCLUSION

In conclusion, the variation of the emission intensity of the CsPbBr$_3$ perovskite nanocrystals in the presence of La$_{0.67}$Sr$_{0.33}$MnO$_3$ and La$_{0.67}$Sr$_{0.33}$Mn$_3$O$_5$ was investigated. These compounds have near room temperature ferromagnetic–paramagnetic phase transition points, and the intensity of the magnetic field can be controlled by varying the temperature. The emission intensity of the CsPbBr$_3$ perovskites is found to increase by 51% in the case of Ni doped La$_{0.67}$Sr$_{0.33}$MnO$_3$ and 33% in the presence of Co doped La$_{0.67}$Sr$_{0.33}$MnO$_3$ on increasing the temperature from 10 $^\circ$C to 35 $^\circ$C. Upon increasing the temperature, the intensity of the magnetic field decreases as a result of the ferromagnetic–paramagnetic phase transition, and this results in the increase in emission from the CsPbBr$_3$ perovskite nanocrystals. Both the magnetic materials and the CsPbBr$_3$ nanocrystals possess perovskite crystal structure, and this might be playing a crucial role in transmitting the magnetic field. These results demonstrate that the thermally varying magnetic field of Ni/Co doped La$_{0.67}$Sr$_{0.33}$MnO$_3$ can be used to control the emission intensity of the CsPbBr$_3$ perovskite nanocrystals and these findings can be used to manipulate the emission intensity in display devices and LEDs. These observations clearly indicate that the magnetic field has a crucial role in controlling the properties of CsPbBr$_3$ perovskites, and these insights can be used for the design of light harvesting devices with better efficiency.

SUPPLEMENTARY MATERIAL

The details of the synthesis characterization of CsPbBr$_3$ perovskites and Ni/Co doped La$_{0.67}$Sr$_{0.33}$MnO$_3$ are given in the supplementary material.

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