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Magnetically Induced Near-Infrared Circularly Polarized Electroluminescence from an Achiral Perovskite Light-Emitting Diode

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Abstract: Circularly polarized electroluminescent devices are conventionally fabricated by incorporating an optically active chiral luminophore into their emission layer. Herein, we developed a circularly polarized perovskite light-emitting diode (PeLED) system with an optically inactive perovskite luminophore that can emit near-infrared circularly polarized electroluminescence (CPEL) upon application of an external magnetic field. The magnitude of the magnetic CPEL (g_{MCPEL}) was in the order of 10^{-3} in the near-infrared wavelength range of 771–773 nm. Although the Pb perovskite quantum dots were achiral, the rotation direction of the CPEL of the magnetic circularly polarized PeLED system was successfully reversed by switching the Faraday geometry of the applied magnetic field. The use of achiral luminophores exhibiting magnetic-field-induced CPEL represents a new approach for the development of circularly polarized electroluminescent devices.

Keywords: chiral; circularly polarized luminescence; circularly polarized perovskite light-emitting diode; circularly polarized organic light-emitting diode; near-infrared circularly polarized electroluminescence; perovskite quantum dot



Citation: Imai, Y.; Amasaki, R.; Yanagibashi, Y.; Suzuki, S.; Shikura, R.; Yagi, S. Magnetically Induced Near-Infrared Circularly Polarized Electroluminescence from an Achiral Perovskite Light-Emitting Diode. *Magnetochemistry* **2024**, *10*, 39. <https://doi.org/10.3390/magnetochemistry10060039>

Academic Editor: Fabrice Pointillart

Received: 8 April 2024

Revised: 22 May 2024

Accepted: 24 May 2024

Published: 28 May 2024



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1. Introduction

Circularly polarized light is a type of polarized electromagnetic wave oscillating in left- and right-handed spirals. In addition to scientific interest in its uniqueness, circularly polarized light has attracted technological interest owing to its applications in three-dimensional displays [1,2], optical communication [3], and other fields [4]. For such applications, electronic devices serving as sources of circularly polarized light are in great demand. In this regard, circularly polarized electroluminescent devices have been developed by doping their emission layer with an optically active chiral luminophore with a high photoluminescence quantum yield (Φ_{PL}) and a high anisotropy coefficient (g_{CPL}) [5–14]. However, these devices have unsatisfactory circular polarization properties and external quantum efficiency; thus, the development of new types of chiral luminophores is eagerly anticipated for the fabrication of such electroluminescent devices.

As a new approach, we have been focusing on the magneto-optical effect on a luminophore in the excited state and previously reported that the application of an external magnetic field to organic light-emitting diodes (OLEDs) leads to the generation of circularly polarized electroluminescence (CPEL) [15–17]. As representative examples, we have developed magnetic circularly polarized OLED systems by incorporating optically inactive, racemic phosphorescent bis- and tris-cyclometalated iridium(III) complexes as emitting dopants [15]. Doping various iridium(III) luminophores into the emitting layer

allowed us to obtain red–green–blue–yellow full-color, circularly polarized electroluminescent devices [16]. Furthermore, employing an excimer-emissive heteroleptic cyclometalated platinum(II) complex as a single emitting dopant allowed us to obtain multi-color CPEL under a magnetic field, where the electroluminescence (EL) intensity ratio of the monomer- and excimer-based emissions was tuned by the doping level of the platinum(II) luminophore [17]. The induction of circularly polarized luminescence (CPL) using such a physical stimulus is an attractive method because optically active luminescent materials are generally difficult to obtain in large quantities owing to the time-consuming optical resolution of racemic products and asymmetric synthetic procedures.

In general, thin-film-based electroluminescent and photovoltaic devices are driven by opposite mechanisms to each other: the former involves the recombination of positive and negative charge carriers in the emitting layer to generate photons from a luminescent material, whereas the latter absorbs photons to induce charge separation in the photo-active layer, leading to an electric potential gap between an anode and a cathode. Metal halide perovskite semiconductors have been actively studied for practical use as raw materials for solar cells [18–24]. They have also attracted attention as main constituents for low-cost, highly efficient, and flexible light-emitting devices because they allow the fabrication of high-quality luminescent thin films with excellent semiconductivity at relatively low temperatures using a simple coating process [25–31]. Recently, chiral perovskite quantum dots capable of exhibiting circularly polarized luminescence (CPL) have been reported [32].

Recently, we successfully induced CPL in achiral inorganic luminescent materials, namely $\text{Y}_2\text{O}_3/\text{Eu(III)}$ and Pb perovskite ($\text{CH}_5\text{N}_2\text{PbBr}_3$) quantum dots (QDs), by applying an external magnetic field of approximately 1.7 T at 25 °C in the solid state or in dilute solution [33,34]. Additionally, three achiral antiferromagnetic Pb perovskite QDs (CsPbCl_3 , CsPbBr_3 , and CsPbI_3) containing different halogen ions emitted full-color red–green–blue magnetic CPL in solution at 25 °C when a 1.7 T external magnetic field was applied. Furthermore, by controlling the combination of halogen ions and their composition ratios, the magnetic CPL color was successfully controlled in solution under a 1.7 T external magnetic field at 25 °C. Notably, the magnetic circularly polarized (MCP) spectral sign could be controlled based on the N-up and S-up directions of Faraday geometry when a 1.7 T external magnetic field was applied.

In a recent report, CPEL was successfully generated from a metal–halide–perovskite (MHP)-based light-emitting diode possessing a chiral MHP hole-transporting layer (so-called “spin-LED”) [35]. This was achieved by injecting spin-polarized charge carriers into the achiral MHP-emitting layer to form spin-polarized carrier pairs. As an alternative method, we attempted to obtain CPL with high brightness in the near-infrared region by applying an external magnetic field to electroluminescent devices based on optically inactive achiral Pb perovskite QDs. Near-infrared light ($\lambda > 700 \text{ nm}$) has useful properties, including invisibility to the naked eye and the ability to penetrate living tissues, for a wide range of technological fields such as night vision [1,36], optical tomography [37,38], and bioimaging [39,40]. Thus, CPL in the near-infrared region has promising applications in security and optical communication. In this study, we developed external-magnetic-field-driven perovskite light-emitting diodes (PeLEDs) capable of emitting near-infrared CPEL by employing an emitting layer consisting of optically inactive achiral Pb perovskite QDs [$\text{Cs}_5(\text{MA}_{0.17}\text{FA}_{0.83})_{95}\text{Pb}(\text{I}_{0.83}\text{Br}_{0.17})_3$; MA = methylamine hydroiodide; FA = formamidinium hydroiodide] [39,40]. Their performance was evaluated by measuring their EL properties.

2. Materials and Methods

2.1. Fabrication of Magnetic Circularly Polarized Electroluminescent Devices

Prepatterned indium tin oxide (ITO) substrates were ultrasonically cleaned in a solution of ultrapure water and alkaline detergent. The surfaces were treated with isopropyl alcohol vapor followed by ultraviolet–ozone treatment. The perovskite layer was then deposited by spin coating, whereas the Ag layer and thin films other than the perovskite layer were deposited by vacuum evaporation. During deposition, the temperature of the

deposition source was monitored, and the deposition rate was precisely adjusted. The deposition system was connected to a glove box (oxygen and moisture concentrations < 10 ppm) via a load lock chamber. After deposition, the devices were placed in a sealed glass container to prevent exposure to the atmosphere.

2.2. Measurement of Electroluminescence (EL), Magnetic Electroluminescence (MEL), and Magnetic Circularly Polarized Electroluminescence (MCPEL)

The MCPEL and unpolarized MEL spectra of the MCP-PeLEDs were acquired at 25 °C using a JASCO CPL-300 spectrofluoropolarimeter (Hachioji, Tokyo, Japan). A 1.7 T external magnetic field was applied using a JASCO PM-491 permanent magnet. The emission bandwidth was 10 nm. The brightness and radiance of the out-coupled EL were recorded using a BM-9 luminance meter (Topcon Technohouse Corporation, Tokyo, Japan) and an SR-NIR near-infrared spectroradiometer (Topcon Technohouse Corporation, Japan), respectively.

3. Results

Two types of MCP-PeLEDs (device I and device II) containing $\text{Cs}_5(\text{MA}_{0.17}\text{FA}_{0.83})_{95}\text{Pb}(\text{I}_{0.83}\text{Br}_{0.17})_3$ as the emitting perovskite layer were fabricated. The devices have the following structure (Figure 1): ITO (cathode, 150 nm)/ SnO_2 (65 nm)/perovskite (548 nm)/2,2',7,7'-tetra(*N,N*-di-*p*-tolyl)amino-9,9-spirobifluorene (spiro-TTB) (20 nm)/ MoO_3 (*x* nm)/Ag (anode, 200 nm). In this study, ITO was used as a transparent cathode. A SnO_2 thin film, a well-known transparent n-type semiconductor, was used as an electron injection layer. Spiro-TTB and MoO_3 were used as hole-transporting and hole-injection layers, respectively. The thickness of the MoO_3 layer (*x*) in devices I and II was adjusted to 10 and 40 nm, respectively.

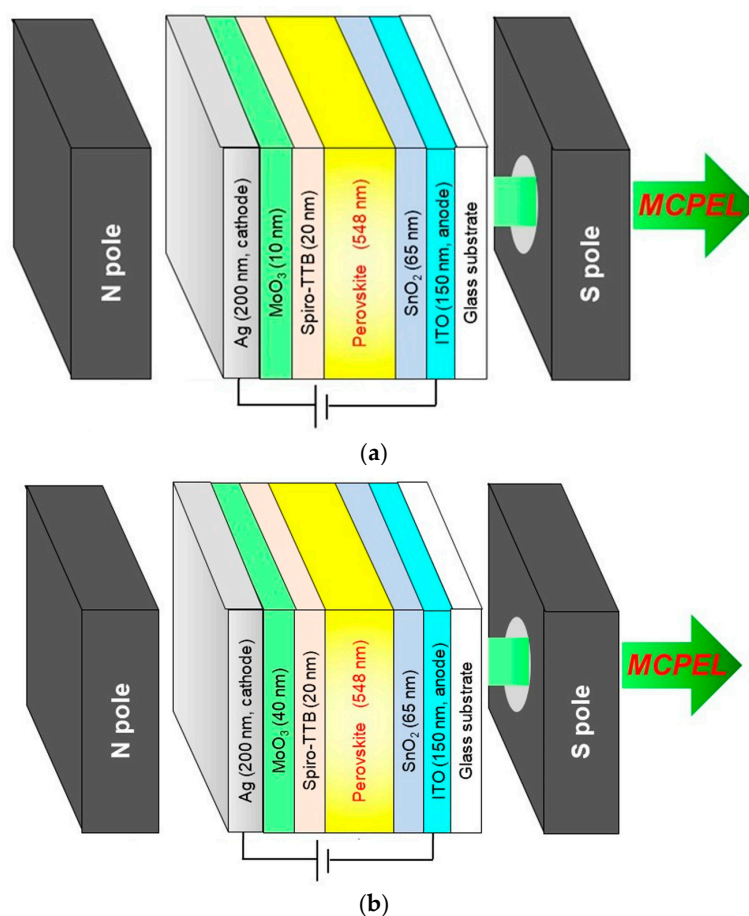


Figure 1. Magnetic circularly polarized electroluminescent devices with permanent magnetic N and S poles. (a) Device I and (b) device II. The values in parentheses are the layer thicknesses. N-up on the diagram.

First, we investigated whether the devices doped with the optically inactive perovskite luminophore could operate as high-performance PeLEDs. In Figure 2, the photographs of devices I and II upon application of an electric voltage of 1.0 V, both of which were obtained using a near-infrared-responsive digital camera equipped with a 700 nm long pass filter, are shown. As expected, these devices emitted near-infrared EL, which was carefully monitored by the present optical recording system, although the out-coupled EL was hardly visible to the naked eye.



Figure 2. Photographs of the electroluminescence of (a) device I and (b) device II at 1.0 V. The cut-off wavelength is 700 nm.

The EL spectra and current density–voltage curves of both devices are shown in Figure 3, and the EL wavelengths (λ_{EL}) and radiances (R) at 1.0 V are summarized in Table 1. Devices I and II exhibited sharp near-infrared EL at 770 and 767 nm, respectively (Figure 3). This indicates that the thickness of the MoO_3 layer does not significantly affect the EL spectral profiles such as the λ_{EL} and the spectral shape. In the current density–voltage profile, the current density of device II was approximately 1.8 times larger than that of device I at any voltage greater than 1.0 V. The values of R for devices I and II were 0.996 and 1.87 $\text{W sr}^{-1} \text{m}^{-2}$, respectively, upon the application of an electric voltage of 1.0 V. This 2-fold difference is consistent with the 2-fold difference in the current density. Thus, the thickness of the MoO_3 layer has a significant effect on the radiance (R) of the device. Specifically, in device II, more charge carriers were injected, which is thought to have resulted in the production of more excitons. These results indicate that the perovskite-based MCP-PeLEDs are suitable for use as near-infrared CPEL devices.

Table 1. Electroluminescence characteristics of devices I and II.

Device	I	II
λ_{EL} (nm)	770	767
R ($\text{W sr}^{-1} \text{m}^{-2}$) [@1.0 V]	0.996	1.87

As expected from the EL spectra, the EL characteristics of devices I and II were sufficient to obtain MEL and MCPeL data. In addition, the MCPeL and MEL properties of these MCP-PeLED devices were investigated at room temperature under a magnetic field (H_0) of 1.7 T, where the electric current was fixed at 10 mA cm^{-2} . The MCPeL and MEL spectra are shown in Figure 4, and the MCPeL properties are listed in Table 2. Both devices exhibited near-infrared MCPeL corresponding to MEL. Notably, mirror-symmetric MCPeL spectra were obtained by alternating the N-up and S-up Faraday arrangements. In other words, the sign of the MCPeL spectrum could be completely controlled by changing the arrangement of the N and S magnets.

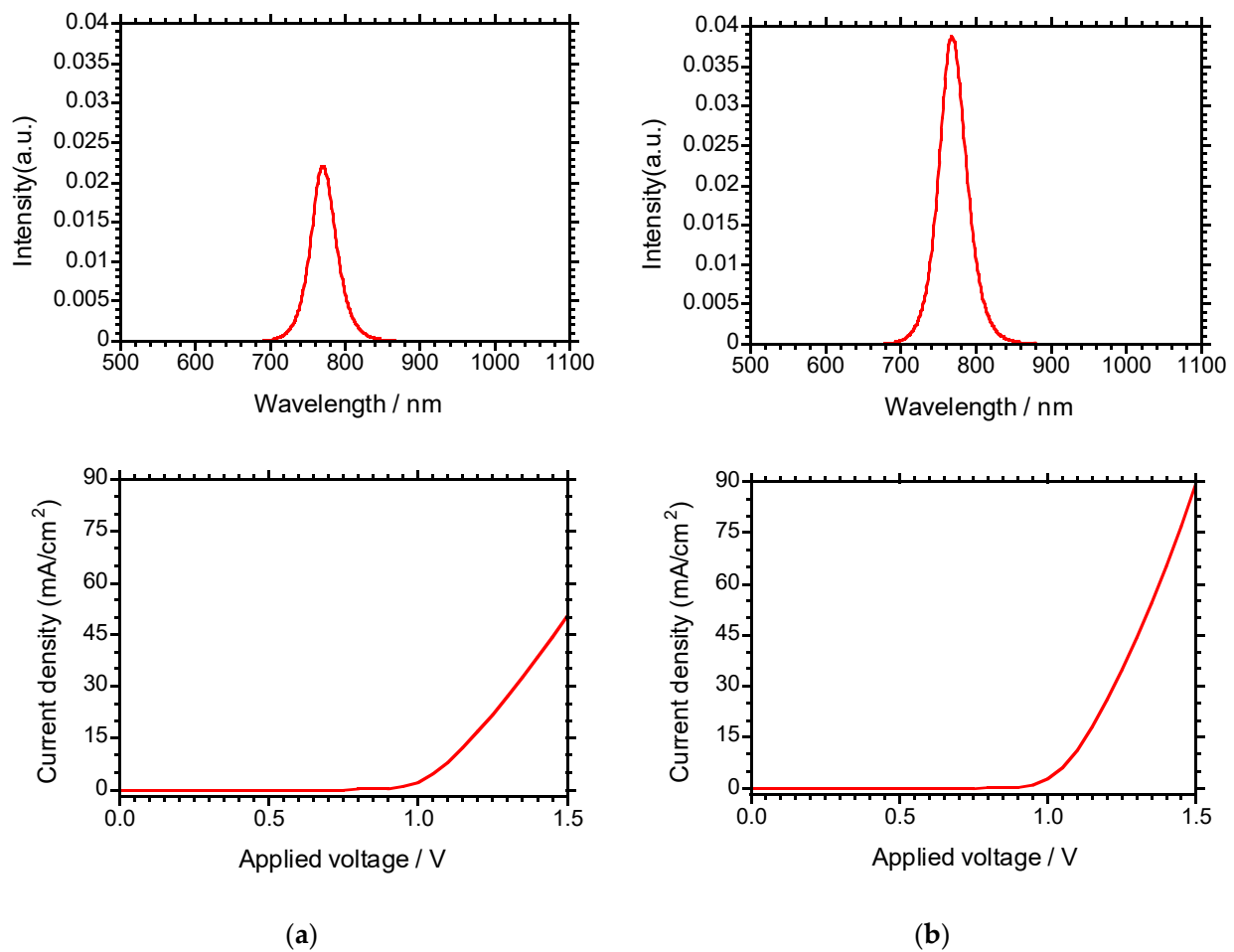


Figure 3. Electroluminescence spectra at 1.0 V (**top**) and current density–voltage profiles (**bottom**) of (a) device I and (b) device II.

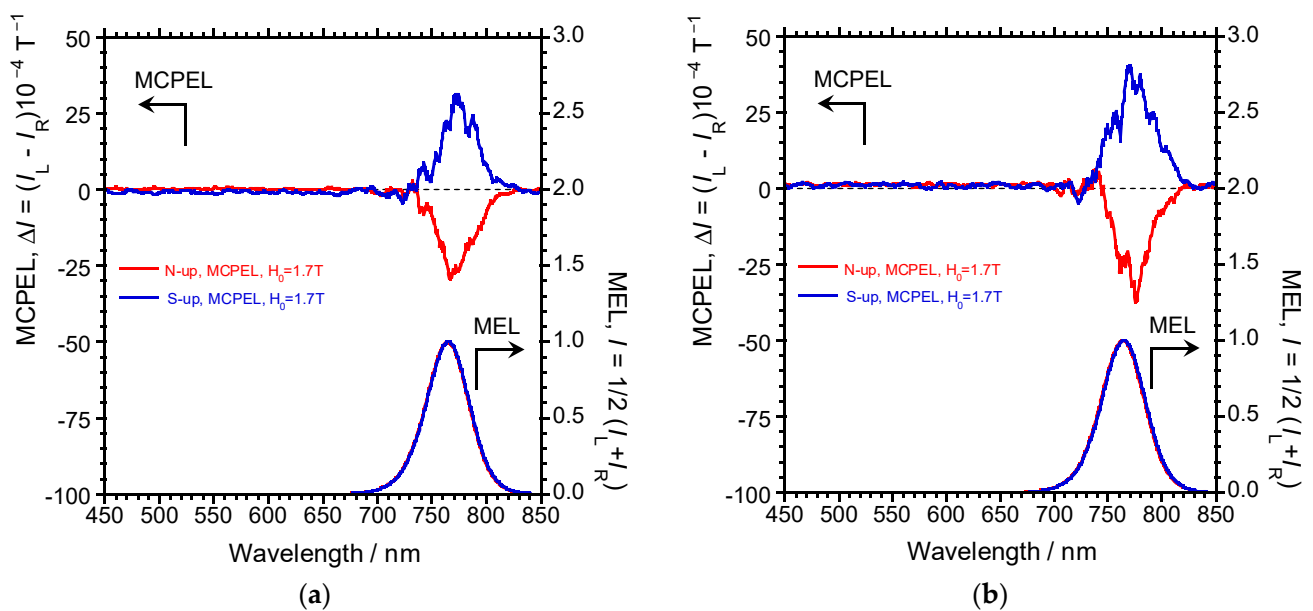


Figure 4. Magnetic circularly polarized electroluminescence (MCPEL, **top**) and magnetic electroluminescence (MEL, **bottom**) spectra at 10 mA cm^{−2} of (a) device I and (b) device II under 1.7 T N-up (red lines) and S-up (blue lines) magnetic fields. I_L and I_R are the amplitude intensities of the left- and right-handed MCPEL, respectively.

Table 2. Magnetic circularly polarized electroluminescence (MCPEL) characteristics of devices I and II under a 1.7 T magnetic field at 10 mA cm^{−2}.

Device	λ_{MCPEL} (nm)	$ g_{\text{MCPEL}} (\times 10^{-3})$ (T ^{−1})	MCPEL Sign for N-Up
I	771	3.3	(−)
II	773	4.3	(−)

For both devices, the MCPEL spectrum of the N-up geometry was negative, whereas that of the S-up geometry was positive. This trend in the MCPEL chiroptical signs is the same as that in the magnetic CPL of Pb perovskite QDs (CH₅N₂PbBr₃) under the same Faraday configuration [34]. In LEDs, when a magnetic field is applied, the orbital of the excited state splits and the degeneracy is lifted due to the Zeeman effect. This is thought to cause a bias in the distribution of electron spins, resulting in circularly polarized light from the LED.

The performance of the MCP-OLED devices was quantitatively evaluated using the dissymmetry factor, $g_{\text{MCPEL}} = (I_L - I_R)/[1/2(I_L + I_R)]$, where I_L and I_R are the amplitude intensities of the left- and right-handed MCPEL, respectively. The MCPEL efficiency, $|g_{\text{MCPEL}}|$, of device I was 3.3×10^{-3} (T^{−1}). Thus, near-infrared MCPEL can be efficiently emitted under an external magnetic field of 1.7 T, even though the employed electroluminescent device contains an optically inactive perovskite luminophore. The $|g_{\text{MCPEL}}|$ of device II was also in the order of 10^{-3} (4.3×10^{-3} (T^{−1})); hence, the difference in g_{MCPEL} was not as dramatic as the 2-fold difference in the radiance values of devices I and II. This indicates that the thickness of the MoO₃ hole-injection layer in the device has a significant effect on exciton generation, but not on the circular polarization properties.

These results indicate that near-infrared circularly polarized emission can be generated from achiral optically inactive perovskite QD semiconductors through hole–electron recombination upon application of an external magnetic field. This near-infrared CPEL system technology using magnetic fields can contribute toward establishing rotation-direction-controllable light sources of near-infrared CPL.

4. Conclusions

We developed external-magnetic-field-driven PeLED devices emitting near-infrared CPEL using an achiral optically inactive perovskite as the emission layer. The chiroptical sign of the MCPEL was completely controlled by the Faraday geometry of the magnetic field. These results provide a new approach for the development of near-infrared CPL materials and near-infrared circularly polarized OLED devices.

Author Contributions: Conceptualization, Y.I.; Investigation, R.A., S.S., Y.Y. and R.S.; Writing—Original Draft Preparation, Y.I.; Writing—Review and Editing, S.Y.; Supervision, Y.I. and S.Y. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by JST-A-STEP [grant number JPMJTM22D9], JST-CREST [grant number JPMJCR2001], Grants-in-Aid for Scientific Research (KAKENHI) [grant numbers JP21K18940, JP23H02040] from MEXT/Japan Society for the Promotion of Science, and grants from the Takahashi Industrial and Economic Research Foundation (2020), Research Foundation for Opto-Science and Technology (2020-5), Ichimura Foundation for New Technology (2020-04), and SCAT Foundation (2021).

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflicts of interest.

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