Reversible multiplexing for optical information recording, erasing, and reading-out in photochromic BaMgSiO₄:Bi³⁺ luminescence ceramics

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ABSTRACT Optical data storage technology has many advantages over the traditional solid-state and magnetic storage technology, such as low cost, multi-dimensional storage, and rewritable capability. Therefore, the optical data storage technology has been in increasing demand for optical storage media. Herein, the photochromic and photoluminescence properties of BaMgSiO₄:Bi³⁺ ceramics were investigated. The BaMgSiO₄:Bi³⁺ ceramics showed reversible photochromism from gray to pink upon alternating the 254 nm ultraviolet light and 532 nm laser irradiation. This is caused by the electron trapping and de-trapping in the oxygen vacancies of the BaMgSiO₄:Bi³⁺ host. This reversible behavior of photochromism was applied to fabricate different patterns on the surface of the BaMgSiO₄:Bi³⁺ ceramics, which exhibited the reversible dual-mode optical information recording and erasing abilities. The photoluminescence reversible modulation of the BaMgSiO₄:Bi³⁺ ceramics was obtained through the photochromic phenomenon. This modification behavior of luminescence could be applied to read-out the recording information in the BaMgSiO₄:Bi³⁺ ceramics. The coloration and bleaching of BaMgSiO₄:Bi³⁺ ceramics were dependent on the time of light stimulation, which facilitated multiplexing encoding. This photoluminescence and photochromism multiplexing of the BaMgSiO₄:Bi³⁺ ceramics enhanced the optical data storage capability.

Keywords: BaMgSiO₄:Bi³⁺ ceramic, photochromism, photoluminescence, reversible modification

INTRODUCTION

With the digitizing and exploding field of information data, the storage life, storage density, and access rate of information data are significantly improved. Optical data

storage technology has many advantages such as high storage density, low cost, long storage life, and non-contact writing and reading capabilities. In contrast to the traditional solid-state and magnetic storage technology, these advantages have increased the application of optical data-storage technology for optical storage media [1–6]. Despite the tremendous progress, the optical storage technology is challenged by storage capacity because of the limitation of optical diffraction [7,8]. To improve the storage capacity, multiplexing the information storage is required [9–12]. Photochromism is always accompanied by the reversible change of chemical substances between two states upon light stimulation, which can switch the absorption property of materials [13–20]. The “0” or “1” digital code can be represented by each state, corresponding to “off” or “on” states. These “off” and “on” characteristics of photochromism promote the potential applications in many fields such as high-density optical memory and optical switches [21–26]. Moreover, the photochromic property was dependent on the time of light stimulation, facilitating multiplexed encoding [27–29]. Thus, the information-storage capacity could be improved based on the photochromic reaction. At present, various organic photochromic optical storage materials such as diarylethene derivatives have been successfully developed [1,30–35]. However, due to their poor thermal reversibility and fatigue resistance, the use of most diarylethenes as the optical storage materials is challenging [36–39]. Therefore, suitable photochromic materials must be investigated to facilitate their application for optical storage.

In contrast to organic photochromic materials, inorganic photochromic materials have many advantages

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such as excellent mechanical strength and chemical and thermal stabilities [40,41]. Various inorganic photochromic materials such as metal oxides (MoO_3 , WO_3 , and V_2O_5), ferroelectrics ($\text{Na}_{0.5}\text{Bi}_{2.5}\text{Nb}_2\text{O}_9$ and $\text{Bi}_4\text{Ti}_3\text{O}_{12}$), and other robust oxides (Mg_2SnO_4 and Sr_2SnO_4) have been developed [15,42–46]. Among inorganic photochromic materials, metal oxides suffer from a slow response time and a poor reversibility [25,47]. Ferroelectrics and robust oxides are more suitable as optical storage materials because of their relatively fast response time and good reversibility. However, thermal bleaching may be required for most of the ferroelectrics and robust oxides such as $\text{Na}_{0.5}\text{Bi}_{2.5}\text{Nb}_2\text{O}_9$, $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$, Mg_2SnO_4 , Sr_2SnO_4 , and $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ [15,48,49]. Light-induced coloring and bleaching are particularly attractive in actual applications. Therefore, development of inorganic photochromic materials with the appropriate light-induced coloring and bleaching properties is necessary for the realization of their optical storage applications.

Inorganic materials doped with rare-earth ions exhibit unique luminescence property. This property can be reversibly modified based on their photochromic reaction [14–16,44]. Different luminescence intensities provide an interesting opportunity to enhance the optical storage potential of the photochromic luminescent materials [50]. Recently, the bismuth ion was extensively used as an active luminescence center [51–55]. In contrast to the rare-earth ions, the bismuth ion has many advantages such as low cost. However, to the best of our knowledge, no reversible luminescence modification of the bismuth-ion-doped phosphors has been reported. BaMgSiO_4 has the tridymite crystal structure with a $P6_3$ space group. The three-dimensional crystal structure of BaMgSiO_4 is connected by the SiO_4 tetrahedrons by sharing their corners. Recently, the photoluminescence of rare-earth-ion-doped BaMgSiO_4 , exhibiting the white light emission or phosphorescence properties, was investigated [56,57]. In this study, $\text{BaMgSiO}_4:\text{Bi}^{3+}$ ceramics were prepared. The photochromic-induced photoluminescence reversible modulation of $\text{BaMgSiO}_4:\text{Bi}^{3+}$ ceramics was investigated in detail. This reversible modification behavior of photochromism and luminescence was applied to fabricate the patterns on the surface of the $\text{BaMgSiO}_4:\text{Bi}^{3+}$ ceramics. These ceramics exhibited the multiplexing ability of reversible optical information recording, erasing, and reading-out.

EXPERIMENTAL

$\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ($x=0.5, 0.75, 1, 1.5$, and 2) ceramics were prepared by using magnesium oxide (MgO ,

99.99%), barium carbonate (BaCO_3 , 99%), silicon dioxide (SiO_2 , 99.99%), and bismuth oxide (Bi_2O_3 , 99.99%) as raw materials, and 10% H_3BO_3 was added as the fluxing agent. Stoichiometric raw materials were mixed in an agate mortar. The mixed powder was pressed into disk, placed in a tubular furnace and sintered at 1350, 1300, or 1250°C in a reducing atmosphere (95% $\text{N}_2+5\% \text{H}_2$) for 5 h. After sintering, the diameter of all the $\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ceramic was about 14 mm. The coloration of $\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ceramics was carried out under the 254 nm ultraviolet light irradiation (ZF-2 Shanghai Anting) for different times with a power density of 0.21 W cm^{-2} . The fading of $\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ceramics was studied upon the 532 nm laser (VD-VII DPSS LASER Jilin Changchun) irradiation with a power density of 380 mW cm^{-2} for various times. The spot size of the 532 nm laser irradiation is $\sim 5 \text{ mm}$ in diameter.

The crystallinity and phase structure of the $\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ceramics prepared at different temperatures were investigated by the X-ray powder diffraction (XRD, Bruker D8 ADVANCE). Field emission scanning electron microscopy (FE-SEM) was used to observe the morphology of the $\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ceramics (QUANTA 650). The thermoluminescence (TL) spectra of ceramics irradiated by 254 nm ultraviolet light for various times were measured by an FJ-427A1 thermoluminescent meter at a heating rate of 1°C s^{-1} . The U-4100 spectrophotometer with an integrated sphere was used to measure the reflectance spectra of $\text{BaMgSiO}_4:\text{Bi}^{3+}$ ceramics. The spectrophotometer (HITACHI U-F-7000) was used to measure the emission and excitation spectra of $\text{BaMgSiO}_4:\text{Bi}^{3+}$ ceramics. The electron paramagnetic resonance (EPR) spectrometer (Bruker X-band A300-6/1) was used to measure the oxygen vacancies in the $\text{BaMgSiO}_4:\text{Bi}^{3+}$ ceramics.

RESULTS

Fig. 1a presents the XRD patterns of $\text{BaMgSiO}_4:x \text{ mol\% Bi}^{3+}$ ($x=0.5, 0.75, 1, 1.5$ and 2) ceramics prepared at 1350°C. The pure BaMgSiO_4 ceramics are classified by the concentration of Bi^{3+} doping. The main XRD peak shifts gradually to a larger degree with the increasing Bi^{3+} doping concentration, as shown in Fig. 1a. The radii of Bi^{3+} and Ba^{2+} are ~ 1.03 and $1.35 \text{ \AA}$, respectively. The Bi^{3+} may occupy the Ba^{2+} sites in the BaMgSiO_4 host. The redshift of XRD peaks is due to the crystal lattice shrinkage induced by the cation substitution of Ba^{2+} with Bi^{3+} . The excitation spectrum of $\text{BaMgSiO}_4:1 \text{ mol\% Bi}^{3+}$ monitored at 510 nm is shown in Fig. 1b. A broad excitation peak centered at 346 nm,

Figure 1 (a) XRD patterns of BaMgSiO_4 : x mol% Bi^{3+} ($x=0, 0.5, 0.75, 1, 1.5$ and 2) ceramics prepared at 1350°C ; (b) excitation spectrum measured at 510 nm emission of BaMgSiO_4 : 1 mol% Bi^{3+} ; (c) luminescence spectra of BaMgSiO_4 : x mol% Bi^{3+} ($x=0, 0.5, 0.75, 1, 1.5$ and 2) ceramics upon the 365 nm excitation.

ranging from 250 to 450 nm was observed. The broad excitation peak of Bi^{3+} is advantageous for the excitation of BaMgSiO_4 : Bi^{3+} ceramic. The luminescence spectra of BaMgSiO_4 : x mol% Bi^{3+} ($x=0.5, 0.75, 1, 1.5$, and 2) ceramics upon 346 nm excitation are shown in Fig. S1. The luminescence property of the BaMgSiO_4 : Bi^{3+} ceramic has no significant change upon the excitation wavelength of 365 or 346 nm. It can be seen from Fig. 1c that the luminescence intensity first increases with the increase of the doping concentration of Bi^{3+} . When the doping concentration of Bi^{3+} is higher than 1 mol%, the luminescence intensity of BaMgSiO_4 : Bi^{3+} ceramic decreases, due to the high-concentration quenching effect. Considering the excitation wavelength, the optimal doping concentration is 1 mol%. In order to achieve the commercial specifications for the BaMgSiO_4 : Bi^{3+} ceramics, in the next section, the 365 nm UV lamp was applied to

excite the ceramics.

The BaMgSiO_4 : 1 mol% Bi^{3+} ceramics prepared at 1400 , 1350 , 1300 , and 1250°C , are denoted as BMSO-1400, BMSO-1350, BMSO-1300, and BMSO-1250, respectively. Fig. 2a exhibits the XRD patterns of BMSO-1400, BMSO-1350, BMSO-1300, and BMSO-1250. The sintering temperature has an influence on the phase structures of BaMgSiO_4 : 1 mol% Bi^{3+} ceramics. The hexagonal phase BaMgSiO_4 : 1 mol% Bi^{3+} ceramics was obtained, and a small amount of miscellaneous phase appeared in the BMSO-1400. Pure phase of BaMgSiO_4 : 1 mol% Bi^{3+} ceramics in the BMSO-1350, BMSO-1300 and BMSO-1250 ceramics was obtained based on the JCPDS Card 16-0573.

Fig. 2b–d show the surface FE-SEM images of the pure phase BMSO-1350, BMSO-1300, and BMSO-1250 ceramics, with compact structures. The particle sizes of BMSO-1350, BMSO-1300, and BMSO-1250 are 8.75 ,

Figure 2 (a) XRD patterns of BMSO-1400, BMSO-1350, BMSO-1300 and BMSO-1250. FE-SEM images of the surface of BMSO-1350 (b), BMSO-1300 (c), and BMSO-1250 (d).

4.52, and 2.67 μm , respectively. The size of BaMgSiO_4 : 1 mol% Bi^{3+} particles increases with the increase of the sintering temperature.

Fig. 3a–c present the diffuse reflectance spectra and corresponding photos of BMSO-1350, BMSO-1300, and BMSO-1250 before and after 254 nm UV light stimulation for 16 min. The color of BMSO-1350, BMSO-1300, and BMSO-1250 ceramics changed from gray to pink as the sintering temperature increased, and a new absorption band at 520 nm in the region of 450–600 nm was observed upon the 254 nm UV light irradiation for 16 min. With an increase in the sintering temperature, the absorption intensity of 520 nm increased and the pink color deepened, as shown in Fig. 3. Fig. S2 presents the XRD patterns of the pink BMSO-1350, BMSO-1300, and BMSO-1250 ceramics after the photochromism, exhibiting no phase change. The results demonstrate that the photochromism of BMSO-1350, BMSO-1300, and BMSO-1250 ceramics is not due to the phase change. The defects such as V''_{Ba} , V''_{Mg} and V''_{O} can be generated in the BaMgSiO_4 :1 mol% Bi^{3+} ceramics. Fig. 3d presents the EPR spectra of BMSO-1350, BMSO-1300, and BMSO-1250 ceramics, showing an EPR peak located at 1.995 of oxygen vacancies. Fig. 3e exhibits the TL spectra of BMSO-1350, BMSO-1300, and BMSO-1250 upon the 254 nm light irradiation for 16 min, exhibiting two TL peaks at 90 and 140°C. These results further indicate that defects such as oxygen vacancies are indeed formed in the BaMgSiO_4 : 1 mol% Bi^{3+} ceramics. Fig. 3f presents the coloration

mechanism of BaMgSiO_4 :1 mol% Bi^{3+} ceramics. The electrons located at the valence band in the BaMgSiO_4 host jump to the conduction band during the 254 nm UV irradiation. The electrons in the conduction band are captured by the oxygen vacancies, and F color centers are formed. Thus, the color change of BaMgSiO_4 :1 mol% Bi^{3+} ceramics from gray to pink is observed. As exhibited in Fig. 3d, e, the TL and EPR intensities increase with the increase of the sintering temperature, indicating that more defects are formed at the high temperature. Thus, better photochromic properties are achieved in the BMSO-1350 ceramic prepared at 1350°C. However, the photochromic property decreases when the temperature increases to 1400°C as shown in Fig. S3, which may be due to the impure phase.

The influence of the irradiation time on the photochromic properties of BMSO-1350 was also investigated. As shown in Fig. 4a, the color deepens and the absorption intensity at 520 nm increases with an increase in the irradiation time. Fig. 4b illustrates the TL intensity change of BMSO-1350 as a function of the irradiation time. The increase in the TL intensity was observed with prolonged irradiation time (at 254 nm), suggesting that more electrons generated in the conduction band were captured by the defects. Thus, the photochromic enhancement of the BMSO-1350 was obtained at the optimal coloration time of ~16 min. The bleaching of the BMSO-1350 after irradiation (254 nm) for 16 min was investigated upon the irradiation with a 532 nm laser (380 mW cm^{-2}) for var-

Figure 3 Diffuse reflectance spectra and corresponding photos of BMSO-1250 (a), BMSO-1300 (b) and BMSO-1350 (c) before and after 254 nm UV light stimulation for 16 min; (d) the EPR spectra of the BMSO-1350, BMSO-1300 and BMSO-1250 without light irradiation; (e) the TL spectra of the BMSO-1350, BMSO-1300 and BMSO-1250 after the 254 nm UV light irradiation for 16 min; (f) the coloration and discoloration mechanism of the samples.

ious times, as shown in Fig. 4c. The decrease in the 520 nm absorption intensity and the color fading was observed with an increase in the 532 nm laser irradiation time. After the 532 nm laser irradiation for 20 min, the color and absorption intensity returned to the initial state. The 532 nm laser irradiation could result in a release of electrons in the defect levels, which recombined with the holes captured by the defects of V''_{Ba} and V''_{Mg} , as shown in Fig. 3f. Thus, the color of BMSO-1350 ceramic faded. The electron release can be demonstrated by the TL spectra. The pink BMSO-1350 ceramic was irradiated by the 532 nm laser for various times, as shown in Fig. 4d. The TL intensity decreased with prolongation of the 532 nm laser irradiation time, suggesting that the number

of electrons in the defect levels gradually decreased.

The photochromic reversibility of BMSO-1350 was investigated by alternating the irradiation between the 254 nm light for 16 min and the 532 nm laser for 20 min, as shown in Fig. 5. This figure shows a good and reversible photochromic property between gray and pink after seven cycles. This result indicates that the $BaMgSiO_4:Bi^{3+}$ ceramics may be viable as optical storage media. The BMSO-1350 ceramic was covered by a mask with sunflower or heart patterns, then irradiated with the 254 nm light for 16 min. The pink information patterns with gray background formed on the surface of BMSO-1350 ceramic (Process ①), as shown in Fig. 5b. It is noted that when the color of BMSO-1350 ceramic was com-

Figure 4 (a) Diffuse reflectance spectra and corresponding photographs, and (b) TL spectra of the BMSO-1350 upon the 254 nm light irradiation for various times; (c) the diffuse reflectance spectra and corresponding photographs, and (d) TL spectra of the pink BMSO-1350 irradiated with the 532 nm laser for various times.

Figure 5 (a) Diffuse reflectance intensity of 520 nm in the BMSO-1350 ceramic upon alternating irradiation between the 254 nm light and the 532 nm laser as a function of the cycle numbers; (b) optical information recording and erasing of BMSO-1350 ceramic realized by using the 254 nm light and 532 nm laser irradiation, and the luminescence reading of optical information of BMSO-1350 ceramic upon the 365 nm excitation.

pletely changed to pink under the irradiation with 254 nm light for 16 min, the information recording can be performed by using the 532 nm laser to irradiate the surface of $\text{BaMgSiO}_4\text{:Bi}^{3+}$ ceramic with sunflower or heart masks. The information pattern with a pink background formed on the surface of the BMSO-1350 ceramic (Process ②). When the pink region of the patterned ceramic was irradiated by using the 532 nm laser for 20 min, the whole ceramic can be completely recovered to raw gray color, directly erasing the information (Process ③). Similarly, when no photochromic region of BMSO-1350-patterned ceramic was irradiated by 254 nm UV light for 16 min, the color of the whole ceramic changed to pink, erasing the information (Process ④). These results demonstrate that the dual-mode reversible optical information recording and erasing was performed, based on the reversible photochromic behavior of the $\text{BaMgSiO}_4\text{:Bi}^{3+}$ ceramics.

The coloration and bleaching of $\text{BaMgSiO}_4\text{:Bi}^{3+}$ ceramics are dependent on the light stimulation time. Fig. S4a shows the four photochromic pink letters of “KUST” on the surface of the BMSO-1350 ceramic irradiated with the 254 nm light for 4, 8, 12, and 16 min, demonstrating the photochromic multiplexing. A similar multiplexing phenomenon was observed after the 532 nm laser bleaching for 2, 8, 14, and 20 min, as shown in Fig. S4b. The multiplex photochromic and bleaching ability can be realized by changing the irradiation time using the 254 and 532 nm light, which could increase the optical storage capacity. Reversible luminescence information recording and reading-out of the $\text{BaMgSiO}_4\text{:Bi}^{3+}$ ceramics are based on the photochromic-induced reaction of luminescence modification.

The photochromic change from gray to pink of the BMSO-1350 was observed upon the 254 nm light irradiation, and the photochromic property was dependent on the irradiation time, as shown in Fig. 4a. The influence of the photochromic phenomenon on the photoluminescence of the BMSO-1350 ceramic was investigated. Fig. 6a shows the luminescence spectra and the corresponding luminescence color change of the BMSO-1350 ceramic upon the 254 nm light irradiation for 0, 4, 8, 12, and 16 min. Under the 365 nm excitation, the BMSO-1350 ceramics before and after the coloration exhibit a 510 nm luminescence peak of Bi^{3+} . Apparently, the 510 nm luminescence of Bi^{3+} was modified by the photochromic reaction. After the 254 nm UV light-induced photochromism, the 510 nm luminescence of Bi^{3+} decreased with increasing time of the 254 nm light irradiation. This can be also identified by the luminescence

image of BMSO-1350 ceramic, as exhibited in the inset of Fig. 6a. The modification degree of the 510 nm luminescence could be expressed by the ΔR_n value: $\Delta R_n = (R_0 - R_n)/R_0 \times 100\%$, where the R_n and R_0 are the luminescence intensity of the photochromic BMSO-1350 ceramic and the luminescence intensity of the raw BMSO-1350 ceramic, respectively. The ΔR_n dependence on the 254 nm light irradiation time is displayed in Fig. 6b. The maximum modification degree of the 510 nm luminescence is about 65%. The color center was generated in the BMSO-1350 ceramic, as exhibited in the EPR spectra. After the photochromism, a 520 nm absorption band in the region of 450–600 nm was observed, which overlapped with the luminescence of Bi^{3+} . The energy transfer may take place from the Bi^{3+} to the color centers, which can quench the luminescence of Bi^{3+} . The energy transfer mechanism can be characterized by the decay lifetime of Bi^{3+} .

Fig. 6c exhibits the decay curves of the BMSO-1350 ceramic at 510 nm after the 254 nm light irradiation for different times. The decay lifetime of Bi^{3+} has no significant change with an increasing irradiation time, indicating a radiative energy transfer from the Bi^{3+} to the color center. The pink BMSO-1350 ceramic can be completely bleached when irradiated with the 532 nm laser for 20 min, as shown in Fig. 4c. The luminescence of the BMSO-1350 ceramic recovered to its initial state when bleached by 532 nm laser irradiation for 20 min. Fig. S5 exhibits the luminescence spectra of the BMSO-1350 ceramic upon alternation between the 254 nm irradiation for 16 min and 532 nm irradiation for 20 min as a function of cycle numbers. The 510 nm luminescence intensity of Bi^{3+} shows no degradation after seven cycles, and a good reversibility and reproducibility was demonstrated over several cycles. As shown in Fig. 6d, switching the states between “on” and “off” for the Bi^{3+} emission could be obtained.

The reversible behavior of photochromism was applied to fabricate the patterns on the surface of the $\text{BaMgSiO}_4\text{:Bi}^{3+}$ ceramics, as shown in Fig. 5b. The information decoding could be realized by the 365 nm light to excite the BMSO-1350 ceramic with and without the photochromic patterns. The luminescence patterns are due to the emission color contrast in BMSO-1350 ceramic based on the photochromic reaction-induced luminescence modification, as shown in Fig. 5b. When the pink-patterned ceramic was irradiated by using 532 nm laser for 20 min, the whole BMSO-1350 ceramic completely recovered to raw gray, and the luminescence erasing was realized upon the 365 nm excitation (Process ⑤), as shown in Fig. 5b. When the gray patterns on the ceramic surface were ir-

Figure 6 (a) Luminescence spectra and corresponding luminescence color change of the BMSO-1350 ceramic upon the 254 nm light irradiation for different times under the excitation of 365 nm; (b) ΔR , dependence on the 254 nm UV light irradiation time; (c) decay curves of the BMSO-1350 ceramic at 510 nm after the 254 nm light irradiation for different times; (d) 510 nm luminescence intensity of BMSO-1350 ceramic upon altering irradiation between 254 nm UV light and 532 nm laser as a function of the cycle number.

radiated with the 254 nm UV light for 16 min, the entire BMSO-1350 ceramic changed to pink, causing the luminescence information to be erased upon the 365 nm excitation (Process ⑥), as shown in Fig. 5b. In order to demonstrate a proof-of-concept of bit-by-bit optical writing and reading-out, the pink photochromic arrays were formed on the surface of the BMSO-1350 ceramic irradiated with the 254 nm light for 16 min, as shown in Fig. 7a. When the color of BMSO-1350 ceramic completely changed to pink under irradiation with 254 nm light for 16 min, the gray photochromic arrays were formed on the surface of the BMSO-1350 ceramic irradiated by the 532 nm light for 20 min, as shown in Fig. 7a. Under the 365 nm UV light excitation, the pink photochromic points or the pink regions show weak green luminescence, while the gray points or regions emit more intense green light, as shown in Fig. 7a. The recorded information could be read-out “bit-by-bit” due to

Figure 7 (a, left) pink and corresponding luminescence photos formed on the surface of the raw BMSO-1350 ceramic irradiated by the 254 nm light for 16 min; (a, right) the gray arrays and corresponding luminescence photos formed on the surface of the pink BMSO-1350 ceramic irradiated by the 532 nm light for 20 min; (b) optical information read-out under the excitation of 365 nm.

the change in the luminescence intensity. As shown in Fig. 7b, the weak and intense luminescence corresponds to the binary states of “0” and “1”, respectively. These results indicate that the BaMgSiO₄:Bi³⁺ ceramics may be available as an optical storage media. Previous investigations demonstrated that the optical storage dimension and density can be expanded by multiplexing the luminescence intensity through the adjustment of the excitation light power [7,8,58]. Fig. S4 shows the luminescence color change for 4 letters of “KUST” for various coloration and bleaching times on the surface of the BMSO-1350 ceramic, demonstrating the luminescence multiplexing. This luminescence multiplexing of BMSO-1350 ceramic may further increase the optical data storage capacity as a multilevel optical medium.

CONCLUSION

In this study, a group of photochromic BaMgSiO₄:Bi³⁺ ceramics were synthesized and their photoluminescence properties were investigated. The reversible color change between gray and pink was observed in the BaMgSiO₄:Bi³⁺ ceramics by alternating the radiation between 254 nm UV light and 532 nm laser. Based on the reversible photochromic behavior, the pink sunflower and heart patterns can be printed on the ceramic surface by 254 nm UV light; the same inverse patterns with the pink background can also be obtained upon 532 nm laser bleaching. The photoluminescence reversible modulation of the BaMgSiO₄:Bi³⁺ ceramics was realized by the photochromic process, promoting the ability to read-out stored information of the BaMgSiO₄:Bi³⁺ ceramics. The coloration and bleaching of BaMgSiO₄:Bi³⁺ ceramics were dependent on the time of light stimulation, exhibiting the luminescence and photochromic multiplexing. This photoluminescence and photochromic multiplexing of the BaMgSiO₄:Bi³⁺ ceramics may enhance the optical data storage capability.

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Author contributions Yang Z and Yu J supervised the project. Ren Y designed and performed the experiments. Qiu J, Wang Y, Li M, Song Z, Ullah A, and Khan I participated in analyzing and interpreting the data. All authors contributed to the general discussion.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

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基于光致变色效应的 $\text{BaMgSiO}_4\text{:Bi}^{3+}$ 陶瓷的发光性质调控及信息的可逆写入、擦除和读出研究

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摘要 光存储技术与传统固态存储和磁存储相比有许多优点, 如成本低、可重复性存储. 因此, 光存储的需求正在持续增长. 本文研究了铋掺杂的硅酸镁钡陶瓷的光致变色及光致发光现象. 在紫外线 (254 nm) 和 532 nm 激光交替辐照下, 实现了陶瓷在灰色与粉红色之间的可逆转变. 研究证实 $\text{MgSiO}_4\text{:Bi}^{3+}$ 陶瓷的可逆光致变色来源于基质中的氧空位陷阱对电子的俘获和释放. 基于可逆光致变色效应在陶瓷表面制作不同颜色的图案, 实现了光信号的双模式存储和擦除. 同时, 在光致变色过程中实现了光致发光的可逆调控; 通过这种可逆的发光调控行为, 实现存储信息的读出; 此外, 变色和漂白的程度与光照射时间有关, 因此可实现多通路编码存储, 有望提高光存储容量.