Abstract: During the past few decades, the research on persistent luminescent materials has focused mainly on Eu$^{2+}$-doped compounds. However, the yearly number of publications on non-Eu$^{2+}$-based materials has also increased steadily. By now, the number of known persistent phosphors has increased to over 200, of which over 80% are not based on Eu$^{2+}$, but rather, on intrinsic host defects, transition metals (manganese, chromium, copper, etc.) or trivalent rare earths (cerium, terbium, dysprosium, etc.). In this review, we present an overview of these non-Eu$^{2+}$-based persistent luminescent materials and their afterglow properties. We also take a closer look at some remaining challenges, such as the excitability with visible light and the possibility of energy transfer between multiple luminescent centers. Finally, we summarize the necessary elements for a complete description of a persistent luminescent material, in order to allow a more objective comparison of these phosphors.

Keywords: persistent luminescence; long-lasting phosphorescence; rare earths

1. Introduction

In most luminescent materials, the decay of the light emission lasts no longer than a few milliseconds after the end of the excitation. On the contrary, persistent phosphors can continue emitting light for minutes or hours. This phenomenon is used in safety signage, dials and displays and
Since the discovery of SrAl₂O₄:Eu²⁺, Dy³⁺ in 1996 [4], many researchers and publications on persistent luminescent materials have focused on divalent europium as the activating ion. An overview of these materials has been presented in an earlier issue of this journal [5]. However, the number of publications on non-Eu²⁺-doped compounds has also seen a steady increase during the past 15 years (Figure 1). In this way, the number of materials where persistent luminescence has been observed has grown continuously over time. By now, over 200 combinations of host materials and activating ions have been described, of which less than 20% is based on divalent europium. In this review article, we will present an overview of the non-Eu²⁺-doped persistent luminescent compounds and their properties.

Figure 1. Number of papers published on non-Eu²⁺-doped persistent luminescent compounds, according to the Web of Science.

The research on non-Eu²⁺-based persistent luminescent materials is mainly driven by the lack of efficient red persistent phosphors. The broadband emission of Eu²⁺ is strongly dependent on the host material, more precisely, on the nephelauxetic effect (or the centroid shift) and the strength of the crystal field acting on the ion [6]. The combination of both effects leads to the so-called red shift, and the value depends strongly on the composition of the host compound and the local coordination of the europium dopant ion. It is quite common to obtain a blue or green afterglow using oxide hosts, but it is much more difficult to find a suitable host material with sufficient red shift, in order to obtain red (persistent) luminescence. Although there are a number of red emitting Eu²⁺-doped persistent phosphors, such as CaS:Eu [7–9] and Ca₂Si₅N₈:Eu [10,11], the choice is limited and the host lattices are chemically unstable or difficult to prepare. This is especially unfortunate, since red afterglow phosphors are strongly desired for several applications, such as safety signage, paints and, more recently, also, as tracer particles for in vivo medical imaging [3,12–14]. Therefore, many research groups have focused on different luminescent ions in order to obtain an efficient red-emitting persistent phosphor.

The most obvious and popular choice for long-wavelength luminescence is Mn²⁺, known for its typical yellow-to-red emission in octahedral sites [15]. In several compounds, an energy transfer from
Eu²⁺ to Mn²⁺ has been observed, leading to a red afterglow color originating from Mn²⁺, but with a long afterglow time defined by Eu²⁺. Not only red-emitting activators are being explored. Other common choices are the different trivalent rare earth ions such as Ce³⁺ and Tb³⁺. An interesting case is Dy³⁺, which shows a white emission color, due to three different emissions around 480, 575 and 665 nm. Such a white emission is very difficult to obtain with only Eu²⁺ doping. Unfortunately, these ions often require a short (UV) excitation wavelength, making it impossible to charge these persistent phosphors using visible light. Finally, several compounds are known to exhibit an afterglow without the addition of (Co) dopants, purely based on the intrinsic luminescence of the host material.

Until 1996, the majority of persistent luminescent applications was based on ZnS doped with copper and cobalt [4,16]. This material emits a greenish broad-band spectrum centered around 540 nm (Figure 2), which remains visible for several hours after the end of the excitation. However, the afterglow of this material is relatively weak, and it was common to add small amounts of radioactive tritium or promethium in order to sustain the luminescence [16]. Since 1996, this ZnS-based phosphor has been rendered obsolete by Eu²⁺-doped strontium and calcium aluminates exhibiting a much brighter and long-lasting afterglow. Nevertheless, the research into non-Eu²⁺-doped persistent phosphors has continuously increased in the background. An extensive list of these phosphors is presented in the following section. The most important ones (with the largest number of publications) to mention at this point are CaTiO₃:Pr³⁺ (red), Y₂O₂S:Eu³⁺, Ti⁴⁺, Mg²⁺ (red) and CaS:Bi³⁺ (blue).

Figure 2. (a) Green persistent luminescence in a Playmobil® ghost toy based on ZnS:Cu, Co. (b) Afterglow emission spectrum of ZnS:Cu, Co centered around 540 nm.

2. Known Compounds

This section provides an overview of the compounds where persistent luminescence, not based on divalent europium, has been reported. For every combination of host compound and activator, relevant references are indicated in the last column. In the case of energy transfer between two different dopants or luminescent centers, both the sensitizer and the activator are indicated. We use the symbol “>>” for efficient energy transfer and “>” for partial energy transfer, as derived from the emission spectra. For clarity, the materials are divided into four groups: silicates, non-silicate oxides,
non-oxides and glasses. If a property was not mentioned explicitly in the text of the reference, but inferred from it or from a figure, it is put between parentheses.

Only materials with an afterglow longer than a few seconds were taken into account, since only in this case, the effect can be termed persistent luminescence. Some publications on phosphors, often using trivalent rare earth elements as dopants, claim to describe persistent luminescence, but only show an effective decay time on the order of milliseconds. In these cases, probably only the intrinsic decay of the forbidden transition within the rare earth ion is observed. Hence, these compounds and publications are deliberately not included in the tables.

The afterglow durations were taken directly from the mentioned references. However, not all of these were measured in a single, clearly defined way. The most common criterion is the visibility by the naked, dark-adapted eye. Only a few authors use the threshold value of 0.32 mcd/m² (which is about 100 times the sensitivity of the human eye and a value often used in the safety signage industry [17]). In some of the references, e.g., [2,18], the afterglow duration was defined as the time the afterglow was measurable with an IR-sensitive camera (in the case of near-IR emission, one could resort to radiometric units [2]). Therefore, the afterglow durations are only noted in the tables as an indication, for a detailed comparison, we refer to the mentioned references.

Furthermore, the exact excitation conditions (wavelength, duration) are not always clear, although 254 nm is a common excitation wavelength. For details on the excitation conditions, we refer to the mentioned references.

2.1. Silicates

Similarly, as in Eu²⁺-doped compounds, the silicates are used as the host crystal for a large part of the non-Eu²⁺-based persistent phosphors (Table 1). Especially, the alkaline earth aluminum and magnesium silicates have been studied extensively. Some of the longest afterglow times (>5 h) have been observed in rare-earth doped CdSiO₃, although the role of host and self-trapped exciton (STE) luminescence remains the subject of discussion [19,20].

**Table 1.** Known non-Eu²⁺-based persistent luminescent silicates (STE = self-trapped exciton).

| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | Reference |
|---------------|------------|------------------------|--------------------|--------------------|-----------|
| Ca₂Al₂SiO₇    | Ce³⁺       | 400–417 (blue)         | identical          | >1 h               | [21–24]   |
|               | Ce³⁺ >> Mn²⁺ | 550 (yellow)          | identical          | >10 h              | [25]      |
| Ca₃Sr₂Al₂SiO₇ | Ce³⁺ > Tb³⁺ | 386, 483 + 542 + 591 (white) | bluish white | >1 min           | [26]      |
| Sr₂Al₂SiO₇    | Ce³⁺       | 400 (near UV)          | identical          | (>2 min)          | [27]      |
|               | Ce³⁺ > Dy³⁺ | 408, 491 + 573 (white) | (identical)        | ~1 h              | [28]      |
|               | Ce³⁺ > Tb³⁺ | 410, 482 + 543 + 588 (white) | (identical) | (>1 min)         | [29]      |
| CaAl₂SiO₆     | Eu²⁺ > Mn²⁺ | 418, 580 (blue)        | identical          | >1 h              | [30]      |
|               | Mn²⁺       | ?                      |                   | ~20 min           | [31]      |
| CaMgSi₂O₆     | Dy³⁺       | 480 + 575 + 667 (white) | identical          | ~2 h              | [32]      |
|               | Eu²⁺ > Mn²⁺ | 450, 580 + 680 (?)     | identical          | (~30 min)         | [33]      |
|               | Mn²⁺       | 580 + 680 (red)        | 680 nm (red)      | >1 h              | [12,33–36]|
| SrMgSi₂O₆     | Dy³⁺       | 455, 576 (blue)        | identical          | >5 min            | [37]      |
|               | Mn²⁺       | 455, 612 (pink)        | (identical)        | (~15 min)         | [37]      |
Table 1. Cont.

| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | reference |
|---------------|------------|-----------------------|---------------------|-------------------|-----------|
| BaMg$_2$Si$_2$O$_7$ | Ce$^{3+}$ > Mn$^{2+}$ | 408, 680 (red) | (identical) | >2 h | [38] |
| | Eu$^{2+}$ > Mn$^{2+}$ | 400, 630–680 (reddish) | (identical) | >2 min | [39–41] |
| | Mn$^{2+}$ | 630–680 nm (red) | (identical) | >30 min | [38,42] |
| Ca$_2$MgSi$_2$O$_7$ | Dy$^{3+}$ | 480 + 575 + 667 (white) | identical | >3 h | [32,43] |
| SrMgSi$_2$O$_4$ | Dy$^{3+}$ | 441, 480 + 575 + 668 (white) | only Dy$^{3+}$ | ~40 min | [44] |
| Ca$_2$MgSi$_2$O$_5$ | Dy$^{3+}$ | 480 + 575 + 667 (white) | identical | >5 min | [32] |
| Sr$_2$Mg$_2$Si$_2$O$_8$ | Eu$^{2+}$ > Mn$^{2+}$ | 457, 670 (?) | identical | >2 h | [45] |
| SrMgAl$_2$Si$_2$O$_7$ | intrinsic/STE | 426 (blue) | (identical) | (~10 min) | [46] |
| | Dy$^{3+}$ | 426, 484 + 572 + 670 (white) | (identical) | (~10 min) | [46,47] |
| | Pr$^{3+}$ | 426, 488 (greenish) | (identical) | (~10 min) | [46] |
| | Sm$^{3+}$ | 426, 565 + 600 + 650 (red) | (identical) | (~10 min) | [46] |
| | Tb$^{3+}$ | 426, 495 + 542 + 590 (green) | (identical) | (~10 min) | [46] |
| Sr$_2$Zn$_2$Mg$_6$Si$_2$O$_8$ | Eu$^{2+}$ > Mn$^{2+}$ | 450, 580 + 680 (near IR) | identical | ~1 h | [3,48] |
| | intrinsic/STE | 380 + 467 + 560 (?) | ~420 (blue) | ~5 h | [19,20] |
| | Dy$^{3+}$ | 410, 486 + 580 (white) | (identical) | >5 h | [49,50] |
| | Eu$^{3+}$, Mn$^{2+}$ | 587, 610 (orange) | (identical) | >1 h | [51] |
| | Mn$^{2+}$ | 572–578 (orange) | identical | ~1–5 h | [52–55] |
| | Mn$^{2+}$, Tb$^{3+}$ | 486 + 548, 587 (orange) | (identical) | >1 h | [56] |
| | Pb$^{2+}$ | 498 (green) | identical | >2 h | [57] |
| | STE > Dy$^{3+}$ | 420, 480 + 575 (white) | identical | ~5 h | [20] |
| | STE > Eu$^{3+}$ | 420, 615 (red) | identical | ~5 h | [20] |
| | STE > Pr$^{3+}$ | 420, 600 (red) | identical | ~5 h | [20,58] |
| | STE > Sm$^{3+}$ | 420, 565 + 600 (pink) | identical | ~5 h | [20,59] |
| | STE > Tb$^{3+}$ | 420, 485 + 540 (green) | identical | ~5 h | [20] |
| | Tb$^{3+}$ | 495 + 545 + 590 (green) | identical | ? | [60] |
| Lu$_2$SiO$_5$ | Ce$^{3+}$ | 400 + 430 (blue) | (identical) | >3 h | [61,62] |
| Mg$_2$SiO$_4$ | Eu$^{2+}$ > Mn$^{2+}$ | (456), 660–665 (?) | 665 (red) | ~4 h | [25,63] |
| Sr$_2$SiO$_3$ | Mn$^{2+}$ | 650 (red) | (identical) | ~20 min | [64] |
| Sr$_2$SiO$_3$ | Dy$^{3+}$ | 480 + 572 + 664 (white) | identical | ~1 h | [65] |
| Sr$_2$ZnSi$_2$O$_7$ | Eu$^{3+}$ | 617 (red) | identical | >20 s | [67] |
| Zn$_2$SiO$_4$ | Mn$^{2+}$ | ? (green) | ? | (>5 min) | [68,69] |
| BaZrSi$_3$O$_9$ | intrinsic/Ti$^{4+}$ | 460–470 (blue) | identical | >20 s | [70,71] |

2.2. Other Oxides

The oxides make up the majority of persistent luminescent compounds, but compared to the Eu$^{2+}$-based materials, many more host compositions (also those in which Eu$^{2+}$ cannot be stabilized) have been explored (Table 2). Besides the aluminates, also the stannates, titanates and germanates show some interesting properties. The longest afterglow durations have been reached in Ce$^{3+}$-doped CaAl$_4$O$_7$, CaAl$_2$O$_4$, SrAl$_2$O$_4$ and BaAl$_2$O$_4$, all with a blue emission color. An exceptional case is the
near-IR afterglow of Cr\(^{3+}\) in LiGa\(_3\)O\(_8\) and Zn\(_3\)Ga\(_2\)Ge\(_3\)O\(_10\) reported by Pan et al., which could be used for night-vision surveillance or in vivo bio-imaging [2,18,72]. Allix et al. found that the latter compound is a variant of the solid solution, Zn\(_{1+x}\)Ga\(_2–2x\).Ge\(_3\)O\(_4\):Cr\(^{3+}\), for \(x = 0.1\). They report even better afterglow properties for the composition with \(x = 0.1\) [73].

Pan et al. mention an afterglow of over 360 h (several weeks), but it should be noted that there is no agreed definition of the afterglow duration for wavelengths that cannot be detected by the human eye. This makes it difficult to compare the various reported afterglow durations.

### Table 2. Other known non-Eu\(^{2+}\)-based persistent luminescent oxides (STE = self-trapped exciton).

| Host material    | Activators       | Emission maximum (nm)        | Afterglow emission | Afterglow duration | Reference |
|------------------|------------------|------------------------------|--------------------|--------------------|-----------|
| BaAl\(_2\)O\(_4\) | Ce\(^{3+}\)      | 402 + 450 (blue)             | (identical)        | >10 h              | [74]      |
|                  | Ce\(^{3+}\)      | 400 (blue)                   | ~413               | >10 h              | [75–77]   |
|                  | Ce\(^{3+}\) >> Mn\(^{2+}\) | 525 (green)                | (identical)        | >10 h              | [25]      |
|                  | Ce\(^{3+}\) >> Tb\(^{3+}\) | 543 (green)                 | identical          | >10 h              | [75,76]   |
| CaAl\(_2\)O\(_4\) | Dy\(^{3+}\)   | 477 + 491 + 577 + 668 (white) | identical          | >30 min            | [78]      |
|                  | Eu\(^{2+}\) > Mn\(^{2+}\) | 440, 545 (green)            | ~440 (violet)      | (>3 h)             | [79]      |
|                  | Tb\(^{3+}\)     | 493 + 543 + 590 + 621 (green) | identical          | ~1 h               | [75,80]   |
| MgAl\(_2\)O\(_4\) | defects        |                              | 520 (green)        | identical          | ~1 h      | [81]      |
|                  | Ce\(^{3+}\)     | 260, 520, 710 (?             | 520, 710 (?)       | (>2 h)             | [82]      |
|                  | Th\(^{3+}\)     | ?                            | ?                  | ~1 h               | [83]      |
| SrAl\(_2\)O\(_4\) | Ce\(^{3+}\)   | 375–385 + 427 (blue)         | only 385           | >10 h              | [84–86]   |
|                  | Ce\(^{3+}\) >> Mn\(^{2+}\) | 375, 515 (green)           | (identical)        | (~5 h)             | [86]      |
|                  | Eu\(^{2+}\) > Eu\(^{3+}\) | 525, 1530 (green/NIR)     | mainly 525         | ~10 min            | [87]      |
|                  | Eu\(^{2+}\) > Nd\(^{3+}\) | 515, 882 (green/NIR)      | mainly 515         | >15 min            | [88]      |
| CaAl\(_2\)O\(_4\) | Ce\(^{3+}\)   | 325, 420 (blue)             | only 420           | >10 h              | [84]      |
| Sr\(_2\)Al\(_2\)O\(_5\) | Ce\(^{3+}\) | 472 + 511 (blue/green)       | (identical)        | ~10 min            | [89]      |
|                  | Eu\(^{2+}\) + Cr\(^{3+}\) | 490, 693 (blue/red) | mainly 490         | >2 h               | [90–92]   |
|                  | Th\(^{3+}\)     | 542 (green)                  | ~380 (blue)        | ?                  | [93]      |
| Y\(_3\)Al\(_2\)O\(_12\) | Ce\(^{3+}\)  | 525 (yellow)                 | identical          | ~2 min             | [94,95]   |
|                  | Mn\(^{2+}\)     | 580 (yellow-orange)          | 585 (orange)       | ~18 min            | [96]      |
|                  | intrinsic, Pr\(^{3+}\) | 300–460, 490 + 610 | 380, 490 + 610     | ?                  | [97]      |
| CaY\(_2\)Al\(_2\)O\(_7\) | Ce\(^{3+}\)  | 425 (blue)                   | (identical)        | ~min               | [21]      |
| CaO              | Eu\(^{3+}\)     | 594 + 616 (red)              | (orange)           | >2 h               | [98,99]   |
|                  | Th\(^{3+}\)     | 550 (green)                  | (identical)        | ?                  | [100]     |
| Ga\(_2\)O\(_3\) | Cr\(^{3+}\)     | 720 (near IR)                | identical          | >4 h               | [101]     |
| HfO\(_2\)        | intrinsic       | 480 (bluish white)           | identical          | >1 min             | [102,103] |
| Lu\(_2\)O\(_3\)  | Eu\(^{3+}\)     | 611 (red)                    | 583 + 594 + 611    | >3 min             | [104]     |
|                  | Th\(^{3+}\)     | 490 + 550 (green)            | identical          | ~5–7 h             | [105–108] |
| SnO\(_2\)        | Sm\(^{3+}\)     | 567 + 607 + 625 (red)        | identical          | ~40 min            | [109]     |
| SrO              | Eu\(^{3+}\)     | 594 + 616 (orange)           | identical          | >1 h               | [99]      |
|                  | Pb\(^{2+}\)     | 390 (violet)                 | identical          | >1 h               | [99]      |
|                  | Th\(^{3+}\)     | 543 (green)                  | (identical)        | ?                  | [100]     |
| Y\(_2\)O\(_3\)   | Eu\(^{3+}\)     | 612 (red)                    | (identical)        | ~90 min            | [110,111] |
| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | reference |
|---------------|------------|-----------------------|--------------------|-------------------|-----------|
| (Zn,Mg)O     | unknown    | 520 (orange)          | (identical)        | ~10 min           | [112,113] |
| ZrO<sub>2</sub> | Sm<sup>3+</sup> | 570 + 614 (red)     | (identical)        | ~15 min           | [114]    |
|              | Ti<sup>4+</sup> | (353+) 470–500 (blue) | only 470–500       | ~1 h              | [115–119] |
| Ba<sub>4</sub>SnO<sub>4</sub> | Sm<sup>3+</sup> | 580 + 611 + 623 (red) | (identical)        | ~20 min           | [120,121] |
| Ca<sub>2</sub>SnO<sub>4</sub> | Eu<sup>3+</sup> | 585 + 618 + 633 (red) | (identical)        | ~50 min           | [122,123] |
|              | STE        | 410 + 466 (blue)     | (identical)        | ~3 h              | [122]    |
|              | STE >> Eu<sup>3+</sup> | 585 + 618 + 633 (red) | (identical)        | ~100 min          | [122]    |
|              | Sm<sup>3+</sup> | 566 + 609 + 653 (red) | (identical)        | >1–7 h            | [120,124–126] |
|              | Tb<sup>3+</sup> | 435, 483 + 545 (blue/green) | 483+545 (green) | ~3 h              | [127]    |
| Mg<sub>2</sub>SnO<sub>4</sub> | intrinsic | 490–495 (green)    | identical          | ~5 h              | [128–130] |
|              | Mn<sup>2+</sup> | 500 (green)           | identical          | >5 h              | [131]    |
| Sr<sub>2</sub>SnO<sub>4</sub> | Sb<sup>3+</sup> | 550 (yellowish white) | identical          | >2 min            | [132]    |
|              | Sm<sup>3+</sup> | 582 + 624 + 672 (red) | identical          | >1 h              | [120,133–135] |
|              | Tb<sup>3+</sup> | 542 (green)           | (identical)        | ~8 min            | [136]    |
| CaSnO<sub>3</sub> | Pr<sup>3+</sup> | 488 + 541 + 620 + 653 (white) | identical         | >3 h              | [137]    |
|              | Sm<sup>3+</sup> | 566 + 601 + 649 + 716 (red) | (identical)       | ?                 | [138]    |
|              | Tb<sup>3+</sup> | 491 + 545 + 588 + 622 (green) | identical         | ~4 h              | [137,139,140] |
| Sr<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> | Sm<sup>3+</sup> | 580 + 621 + 665 + 735 (red) | identical         | >1 h              | [141]    |
| Ca<sub>4</sub>Gd(PO<sub>4</sub>)<sub>2</sub> | Mn<sup>2+</sup> | 602 + 628, 660 (red) | only 660 (red)    | (>20 min)         | [142]    |
| Ca<sub>4</sub>Lu(PO<sub>4</sub>)<sub>2</sub> | Mn<sup>2+</sup> | 660 (red)            | identical         | (>20 min)        | [142]    |
| Ca<sub>4</sub>Tb(PO<sub>4</sub>)<sub>2</sub> | Tb<sup>3+</sup> | 490 + 545 (green)    | (identical)        | (>20 min)        | [142]    |
| Ca<sub>4</sub>(PO<sub>4</sub>)<sub>2</sub> | Mn<sup>2+</sup> | 645–660 (red)        | identical         | ~1 h             | [143,144] |
| SrMg<sub>2</sub>(PO<sub>4</sub>)<sub>2</sub> | Eu<sup>3+</sup>, Zr<sup>4+</sup> | 500, 588 (white) | (identical)        | ~1.5 h           | [145]    |
| SrZn<sub>2</sub>(PO<sub>4</sub>)<sub>2</sub> | Eu<sup>2+</sup> + Mn<sup>2+</sup> | 421, 547 (white) | (identical)        | ~1 min           | [146]    |
|              | Mn<sup>2+</sup> | 547 (green)          | (identical)        | ~1 min            | [146]    |
| Zn<sub>2</sub>(PO<sub>4</sub>)<sub>2</sub> | Hf<sup>4+</sup> | 470 (blue)           | identical         | >40 min           | [147]    |
|              | Mn<sup>2+</sup> | 616 (red)             | identical         | >2 h              | [148–151] |
|              | Mn<sup>2+</sup>, Zr<sup>4+</sup> | 475, 616 (blue/red) | mainly 616       | ~3 h              | [152]    |
| YPO<sub>4</sub> | Pr<sup>3+</sup> | 600 + 620 (orange/red) | (identical)        | >30 min           | [153]    |
| Ca<sub>4</sub>Mg<sub>2</sub>TiO<sub>3</sub> | Pr<sup>3+</sup> | 613 (red)            | (identical)        | ?                 | [154]    |
| CaTiO<sub>3</sub> | Pr<sup>3+</sup> | 612 (red)            | identical         | >2 h              | [155–159] |
| (Ca,Zn)TiO<sub>3</sub> | Pr<sup>3+</sup> | 612 (red)            | (identical)        | ~20 min           | [160–162] |
| Ca<sub>2</sub>Zn<sub>4</sub>Ti<sub>11</sub>O<sub>30</sub> | Pr<sup>3+</sup> | 614 + 644 (red)     | mainly 614        | ?                 | [163,164] |
| La<sub>4</sub>Ti<sub>17</sub>O<sub>30</sub> | Pr<sup>3+</sup> | 611 (red)            | identical         | >1 h              | [165]    |
| GdGa<sub>5</sub>O<sub>12</sub> | Cr<sup>3+</sup> | 697 + 716 (red)     | (identical)        | ?                 | [166,167] |
| MgGa<sub>2</sub>O<sub>4</sub> | Mn<sup>2+</sup> | 506 (green)          | (identical)        | ?                 | [168]    |
| LiGa<sub>2</sub>O<sub>6</sub> | Cr<sup>3+</sup> | 716 (near IR)       | identical         | >1000 h           | [18]     |
| ZnGa<sub>2</sub>O<sub>4</sub> | defects   | 410 + 540 (white)   | identical         | ~40 min           | [169]    |
|              | Cr<sup>3+</sup> | 650–750 (red)       | identical         | >1 h              | [72,73]  |
|              | Mn<sup>2+</sup> | 504 (green)          | (identical)        | >15 min           | [170]    |
| (Zn,Mg)Ga<sub>2</sub>O<sub>4</sub> | Mn<sup>2+</sup> | 505 (green)         | (identical)        | >15 min           | [170]    |
| Cd<sub>2</sub>Ge<sub>2</sub>O<sub>6</sub> | Mn<sup>2+</sup> | 585 (orange)        | identical         | >3 h              | [171]    |
|              | Pb<sup>2+</sup> | 352 + 497 (blue)    | only 497          | ~10 min           | [172]    |
### Table 2. Cont.

| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | reference |
|---------------|------------|------------------------|--------------------|-------------------|-----------|
| MgGeO$_3$     | Mn$^{2+}$  | 650–670 (red)          | identical          | ~30 min           | [173,174] |
| Zn$_2$GeO$_4$ | Mn$^{2+}$  | 528 (green)            | (identical)        | >2 h              | [175]     |
| CaZnGe$_2$O$_6$| Dy$^{3+}$  | (white)                | (identical)        | >3 h              | [176]     |
|               | Mn$^{2+}$  | 648 (red)              | identical          | >3 h              | [177]     |
|               | Tb$^{3+}$  | 488 + 552 + 583 + 622 (green) | identical | ~4 h | [178,179] |
| Cd$_3$Al$_2$Ge$_4$O$_{12}$ | intrinsic > Dy$^{3+}$ | 437, 485 + 580 (?) | (identical) | ~1 h | [180] |
| La$_3$Ga$_5$Ge$_2$O$_{14}$ | Cr$^{3+}$ | 785, 960–1030 (near IR) | only 960–1030 | >1–8 h | [181,182] |
| Zn$_2$Ga$_2$Ge$_5$O$_{10}$ | Cr$^{3+}$ | 696 + 713 (near IR) | identical | >360 h | [2,73] |
| CaMoO$_4$ | Eu$^{3+}$ | 616 (red)              | identical          | >5 min            | [183]     |
| NaNbO$_3$     | Pr$^{3+}$  | 620 (red)              | identical          | ?                 | [184]     |
| YTaO$_4$      | Tb$^{3+}$  | 492 + 543 + 590 + 624 (green) | (identical) | ~2 h | [185] |
| CaWO$_4$      | intrinsic > Pr$^{3+}$ | 415, 490 + 650 (blue/white) | identical | >10 min | [186] |
|               | Eu$^{3+}$  | 592 + 616 (red)        | identical          | >40 min           | [187–189] |
|               | Sm$^{3+}$  | 592 + 616 (red)        | (identical)        | >35 min           | [190]     |
|               | Tb$^{3+}$  | 490 + 546 (green)      | identical          | (>10 min)         | [191]     |
| BaZrO$_3$     | Eu$^{3+}$  | 574 + 596 + 614 (red)  | (identical)        | (~10 min)        | [193]     |
|               | Ti >> Eu$^{3+}$ | 574 + 596 + 614 (red)  | (identical)        | (~10 min)        | [193]     |

2.3. Other Compounds

The sulfides (Table 3) have the longest recorded history of all persistent luminescent compounds. In fact, the famous Bologna Stone, discovered by Vincenzo Casciarolo in 1602 [194], consisted mainly of copper-doped BaS [195]. Nowadays, the use of ZnS: Cu has much decreased in favor of SrAl$_2$O$_4$:Eu, Dy. The focus has mainly shifted to the oxysulfides, especially Y$_2$O$_2$S:Eu$^{3+}$, Ti$^{4+}$, Mg$^{2+}$, which is currently one of the best red-emitting persistent phosphors. Nevertheless, its afterglow intensity is much weaker than the Eu$^{2+}$-doped aluminates or silicates [196]. An interesting case of persistent luminescence is observed in undoped BCNO, where the emission wavelength can be shifted from blue to orange purely by changing the preparation conditions.

### Table 3. Other known non-Eu$^{2+}$-based persistent luminescent compounds.

| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | reference |
|---------------|------------|------------------------|--------------------|-------------------|-----------|
| BaS           | Cu$^+$     | 610 (orange)           | (identical)        | >30 min           | [195]     |
| CaS           | Bi$^{3+}$  | 448 (blue)             | (identical)        | (~20 min)         | [8,197–199] |
|               | Ce$^{3+}$  | 508 + 568 (green)      | (identical)        | ~5 min            | [200]     |
|               | Sm$^{3+}$  | 569 (green)            | ?                  | (~3 h)            | [201]     |
| (Ca,Sr)S      | Bi$^{3+}$  | 453 (blue)             | (identical)        | (>15 min)         | [202]     |
| SrS           | defects    | 517 (green)            | (identical)        | (~20 min)         | [203]     |
| ZnS           | Cu$^+$     | 530 (green)            | (identical)        | (>3 h)            | [16,69,204,205] |
### Table 3. Cont.

| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | reference |
|---------------|------------|-----------------------|--------------------|--------------------|-----------|
| Gd<sub>2</sub>O<sub>3</sub> | Ti<sup>3+</sup>/defects | 590 (orange) | identical | ~2 h | [206,207] |
| | Ti<sup>3+</sup> > Er<sup>3+</sup> | 555 + 675 (green) | 555 + 675, 590 | >1 h | [206,208] |
| | Ti<sup>3+</sup> >> Eu<sup>3+</sup> | 504 + 536 + 620 (red) | identical | (>5 min) | [206,209,210] |
| | Ti > Sm<sup>3+</sup> | 607 (red) | 590, 607 | ? | [206] |
| | Ti > Tm<sup>3+</sup> | 513 + 800 (?) | 590, 800 | ? | [206] |
| La<sub>2</sub>O<sub>3</sub> | Sm<sup>3+</sup> | 605 + 645 + 656 (red) | (identical) | (>1 min) | [211] |
| Y<sub>2</sub>O<sub>3</sub> | Ti<sup>3+</sup>/defects | 540–594 (orange) | identical | >5 h | [212–216] |
| | Eu<sup>3+</sup> | 590 + 614 + 627 + 710 (red) | identical | ~3 h | [217–220] |
| | Sm<sup>3+</sup> | 570 + 606 + 659 (red) | (identical) | >1 h | [221–223] |
| | Tb<sup>3+</sup> | 417 + 546 (green) | (identical) | >20 min | [224] |
| | Ti<sup>3+</sup> > Eu<sup>3+</sup> | 616 + 625 (red) | 565, 616 + 625 | ~10 min–5 h | [225,226] |
| | Tm<sup>3+</sup> | 495 + 545 + 588 (orange) | identical | ~1 h | [227] |
| BCNO | intrinsic | 520 (green) | identical | >2 h | [228,229] |
| Ba<sub>4</sub>(PO<sub>4</sub>)Cl | Ce<sup>3+</sup> > Eu<sup>2+</sup> | 350, 435 (blue) | only 435 | (>5 min) | [230] |
| KY<sub>2</sub>F<sub>10</sub> | Sm<sup>3+</sup> | 558 + 597 + 651 (red) | (identical) | (>2 min) | [231] |
| ZnSiN<sub>2</sub> | Mn<sup>2+</sup> | 620 (red) | (identical) | ~min | [232] |

### 2.4. Glasses

A final group of persistent luminescent compounds are the glasses (Table 4). Although it is sometimes difficult to accurately infer the composition of these glasses from the publications, some clear trends can be observed. Especially, the calcium aluminum silicate and zinc boron silicate glasses have a long afterglow of more than one hour.

### Table 4. Known non-Eu<sup>2+</sup>-based persistent luminescent glasses.

| Host material | Activators | Emission maximum (nm) | Afterglow emission | Afterglow duration | reference |
|---------------|------------|-----------------------|--------------------|--------------------|-----------|
| Ca<sub>4</sub>Al<sub>6</sub>Si<sub>3</sub>O<sub>19</sub> | Ce<sup>3+</sup> | ? (blue) | (identical) | >1 h | [233] |
| | Pr<sup>3+</sup> | ? (red) | (identical) | >1 h | [233] |
| | Tb<sup>3+</sup> | 350–600 (green) | identical | >1 h | [233] |
| Ca<sub>6</sub>Al<sub>8</sub>Si<sub>3</sub>Mg<sub>3</sub>O<sub>16</sub> | Mn<sup>2+</sup> | 540 (yellow) | (identical) | >1 h | [234] |
| | Pr<sup>3+</sup> | 493 + 610 (red) | (identical) | >1 h | [234] |
| | Tb<sup>3+</sup> | 543 (green) | (identical) | >2 h | [234–236] |
| GeO<sub>2</sub> | intrinsic | 465 (blue) | identical | (~20 min) | [237,238] |
| SiO<sub>2</sub> | defects | 290 + 390 (blue) | identical | ~1 h | [239] |
| Na<sub>2</sub>AlB<sub>2</sub>O<sub>4</sub> | Mn<sup>2+</sup> | 590 (reddish) | identical | ~5 min | [240] |
| Na<sub>2</sub>CaGa<sub>2</sub>Si<sub>3</sub>O<sub>12</sub> | Tb<sup>3+</sup> | 542 (green) | identical | ~1 h | [241] |
| Na<sub>2</sub>CaSi<sub>3</sub>O<sub>12</sub> | Cu<sup>2+</sup>/Cu<sup>2+</sup> | 510 (blue green) | identical | >30 min | [242] |
| Sr<sub>2</sub>B<sub>2</sub>O<sub>4</sub> | Eu<sup>2+</sup>, Ce<sup>3+</sup> | 350, 430 (blue) | mainly 430 | (>2 min) | [243] |
| ZnGe<sub>2</sub>O<sub>4</sub> | Mn<sup>2+</sup> | 534 (green) | identical | >1 h | [244] |
| Zn<sub>2</sub>GeO<sub>4</sub> | Mn<sup>2+</sup> | 540 (green) | identical | (>10 s) | [245] |
Table 4. Cont.

| Host material          | Activators          | Emission maximum (nm)          | Afterglow emission | Afterglow duration | reference       |
|------------------------|---------------------|--------------------------------|---------------------|-------------------|----------------|
| Zn$_2$B$_2$SiO$_8$     | Pr$^{3+}$           | 495 + 603 (reddish)            | identical           | (>30 min)         | [246]          |
|                        | Tb$^{3+}$           | 542 (green)                    | identical           | ~1 h              | [247,248]      |
| Zn$_{11}$B$_8$Si$_5$O$_3$ | Mn$^{2+}$            | 525–606 (green/yellow)       | identical           | ~12 h             | [249]          |
| Zn$_{11}$B$_{10}$Si$_4$O$_{34}$ | Mn$^{2+}$, Sm$^{3+}$ | 590 (red)                      | identical           | (~20 min)         | [250]          |
|                        | Mn$^{2+}$, Yb$^{3+}$ | 600 (red)                      | identical           | ~10 h             | [251,252]      |
|                        | Mn$^{2+}$, Yb$^{3+}$ | 605, 980 (red/IR)             | identical           | (~10 min)         | [253]          |
| Zn$_{60}$B$_{40}$Si$_{17}$Ge$_3$A$_{16}$O$_{160}$ | defects | 410 (blue)                     | identical           | ~2 h              | [254]          |

3. General Remarks

It is very difficult to draw general conclusions from the above tables. One of the most interesting activators is Cr$^{3+}$, which is not commonly used, but shows some excellent afterglow properties as a red/near-IR luminescent center. This might be especially useful for in vivo medical imaging applications. Unfortunately, even though the excitation spectrum of Cr$^{3+}$ for steady-state luminescence extends to about 650 nm, it is very difficult to fill the traps, which are necessary to obtain afterglow, using visible light (about 40 times less efficient compared to UV light) [2] (Figure 3).

3.1. Excitation Difficulties

From Figure 3, it is immediately clear that the steady-state excitation spectrum and afterglow excitation spectrum are not always the same. In many persistent luminescent materials, it is much easier to fill traps using higher energy photons (i.e., using shorter excitation wavelengths) [2,255]. This implies that direct bandgap excitation is much more efficient to fill the traps than excitation of the luminescent centers. Even more problematic, the latter type of excitation might require a certain thermal activation barrier to be surpassed before traps can be filled [255], making the use of visible light even less favorable. Of course, it is also possible to fill the traps directly through tunneling from the activating ions, which does not require short wavelength excitation, but is clearly less efficient. These different trapping processes are shown on an energy level diagram in Figure 4.

This effect appears to be even more profound in non-Eu$^{2+}$-based persistent phosphors, where, in general, only UV light is able to effectively fill the traps in the material. This implies that the role of the host compound is much larger than in Eu$^{2+}$-based materials. While it has been shown that in Eu$^{2+}$-based persistent phosphors, the activator is a main source of trapped electrons [256], in non-Eu$^{2+}$-based compounds, the trapped charge carriers are created mainly after band gap excitation. The luminescent center is subsequently excited by energy transferred from the traps when the trapped electron and hole recombine. The same phenomenon is illustrated by the fact that the afterglow duration is influenced much more by the host compound than by the actual luminescent center. Indeed, by looking at the tables presented in Section 2, it is not uncommon to see certain host compounds with very similar afterglow durations irrespective of the activator being, e.g., Pr$^{3+}$, Sm$^{3+}$ or Tb$^{3+}$.

The fact that UV excitation is required for efficient trap filling is especially unfortunate for persistent phosphors based on Dy$^{3+}$. This could be an excellent activator for white persistent
luminescence, e.g., in paints, signage and displays. However, since indoor lighting contains little to no UV wavelengths (especially with the advent of LED lighting [257]), these compounds are not suited for practical indoor applications.

**Figure 3.** (a) Excitation and emission spectrum of Zn$_3$Ga$_2$Ge$_2$O$_{10}$:0.5%Cr$^{3+}$; (b) Effectiveness of excitation wavelength (energy) for persistent luminescence of Zn$_3$Ga$_2$Ge$_2$O$_{10}$:0.5%Cr$^{3+}$. The afterglow intensity after 10 s is monitored as a function of the excitation wavelength (Reprinted with permission from [2]).

**Figure 4.** Energy level diagram for CaAl$_2$O$_4$: Ce$^{3+}$, showing the positions of the Ce$^{3+}$ levels relative to the bandgap of the host and the proposed trapping mechanism. After excitation in the conduction band, trapping occurs through the conduction band. After excitation in the lower 5d levels, trapping occurs through tunneling (Reprinted with permission from [77]. Copyright 2003 The Electrochemical Society).
3.2. Energy Transfer

In several persistent luminescent compounds, energy transfer has been reported. Two types of energy transfer can be distinguished in this case. The first type is the transfer of excitation energy between a sensitizer and an activator. However, we are more interested in the second type, where energy is transferred during the afterglow phase, after the end of the excitation. When the first activating ion recombines, instead of emitting a photon, it can transfer this recombination energy to a second activating ion. This makes it possible to see or extend the afterglow emission from activators that usually have little to no persistent luminescent properties. If the energy transfer is very efficient, only emission from the second activator, receiving the recombination energy, can be observed. In the other case, luminescence from both kinds of activators can be seen simultaneously in the afterglow spectrum.

It is not always immediately clear if energy transfer is present or not. The afterglow spectrum can consist of the emission of two different kinds of activators, even when no energy is transferred between them. It is therefore necessary to carefully inspect the decay behavior of both kinds of activators. If the decay rates of both are the same, this indicates that one of them is transferring its recombination energy to the other. If no energy transfer is present, it is likely that both kinds of activators will have a (slightly) different decay behavior, and the shape of the afterglow spectrum might change over time.

4. Tools for an Accurate Description of a Persistent Luminescent Material

There is no standard way to describe the properties of a given persistent luminescent material. The multitude of parameters, the uncertainties about the underlying mechanism and the lack of clear definitions make an accurate and complete description or comparison nearly impossible. Ideally, there are certain elements and experiments that should always be addressed in a publication on persistent phosphors. This allows for an easier interpretation of experimental results and simplifies the comparison between different persistent luminescent materials.

The emission and excitation spectrum during fluorescence should be given, in order to know which activators are taking part in the luminescent process. When multiple peaks or bands are present in the excitation or emission spectrum, the corresponding emission and excitation spectra for each peak should be measured. Ideally, an excitation-emission mapping is provided, offering a complete overview of the emission spectrum for every possible excitation wavelength. This can also unveil the presence of energy transfer during the excitation or emission process.

Not only the steady-state emission spectrum during excitation, but also the afterglow emission spectrum should be shown, since these can differ drastically from each other. In this way, it is clear which activators are taking part in the persistent luminescence and which don’t. If the afterglow emission spectrum changes over time, it might be valuable to show the spectrum at different time intervals after the excitation. For applications, it might be useful to mention both the fluorescence and the afterglow color. If multiple luminescent centers are present, a thermoluminescence (TL) experiment, where the emission spectrum is measured (TL-emission mapping, Figure 5), provides information about which traps are connected to which luminescent centers. For example, a certain activator might only emit at higher temperatures, indicating that it is only connected to deeper traps in the material.
Figure 5. Thermoluminescence (TL)-emission mapping: the emission spectrum is monitored during the thermoluminescence experiment, showing which traps are related to which activators. An example is shown for Mn$^{2+}$-emission in CaMgSi$_2$O$_6$ (Reprinted with permission from [33]. Copyright 2010 Elsevier)

If any TL measurements are made, it is advisable to perform an entire series instead of a single experiment, by varying a single parameter and keeping the other parameters constant. These parameters include the duration of the excitation, the excitation intensity, the heating rate and the delay between excitation and the start of the TL experiment (fading time). A TL-excitation mapping (Figure 6)—where the TL experiment is repeated for various excitation wavelengths—is especially useful, since it directly provides information on the trap filling probability of different wavelengths [258]; i.e., it shows which excitation wavelengths are suitable for inducing persistent luminescence. This can then be compared to the steady-state excitation spectrum to see which processes occur during fluorescence and during trap filling.

Figure 6. TL-excitation mapping: the TL measurement is repeated for different excitation wavelengths, showing which wavelengths are suited for trap filling. An example is shown for Cu$^{+}$-emission in ZnS (presented earlier in [258]). It can be seen that different kinds of traps are being filled by short (<340 nm) and longer (>340 nm) wavelengths, where 340 nm corresponds to the band gap of the ZnS host compound.
A further study of the trapping system can be done by performing TL experiments after excitation at different temperatures or by partial thermal emptying of the sample traps before the experiment. In this way, the depth of the various traps can be obtained or the presence of a trap depth distribution can be revealed [259]. Indeed, if such a distribution is present, exciting at higher temperatures or partial thermal cleaning will lead to only deeper traps being filled and shallower traps being emptied. Therefore, the estimated trap depth obtained from a TL experiment will become continuously deeper for higher excitation temperatures, which proves the existence of the trap depth distribution. Under the right conditions, it is even possible to derive the shape of this distribution (Gaussian, uniform, exponential, etc.) [260].

For further study of the defects, one can also turn to electron paramagnetic/spin resonance (EPR/ESR), which provides more information on the structural properties of the defects [24,261].

Finally, it is important to clearly state the exact experimental conditions, such as the dopant and codopant concentrations and the excitation wavelength and duration. If the duration of the afterglow decay is given, information should be given on how this was determined. According to DIN 67510-1, the sample should be excited for five minutes by 1000 lx light of an unfiltered Xe arc lamp. However, the emission spectrum of a Xe lamp is very broad and contains UV, visible, as well as infrared light. This makes it hard to draw conclusions on the excitability, based on such a measurement. It does not give a good prediction of how the persistent luminescent material will behave when excited by artificial light or sunlight. It might be more interesting to excite with monochromatic light at different wavelength and compare the afterglow in each situation.

The afterglow intensity decay should be measured in cd/m², and the afterglow duration should be the time between the end of the excitation and the moment when the afterglow intensity drops below 0.32 mcd/m², a value commonly used by the safety signage industry (about 100-times the sensitivity of the dark-adapted eye [17]). In this way, it would be very simple and straightforward to compare the performance of different persistent luminescent materials. However, this definition is not applicable for UV- or NIR-emitting persistent phosphors, where the luminous emission is zero by default and no clear definition exists for the afterglow duration. In that case, one can resort to radiometric units [2].

Furthermore, such absolute measurements of the afterglow decay could provide important information on the absolute concentration of activators, defects and trapped charge carriers.

5. Conclusions and Perspectives

A lot of research is going on in the field of non-Eu²⁺ persistent luminescent materials, and numerous material-dopant combinations have been and are being developed. However, up to now, the best Eu³⁺-based persistent phosphors are still without competition in terms of absolute luminance and afterglow time, apart from certain Cr³⁺-doped phosphors. Since the process of persistent luminescence is based on a delicate interplay between energy levels of dopants and co-dopants, intrinsic defects and energy bands of the host lattice and the possible physical proximity of dopants and co-dopants, small changes in composition, material purity and crystallinity and dopant concentration can have a strong effect on the afterglow properties. Most probably, the optimum material properties, especially the total amount of stored energy, have not been achieved yet, and most likely, some of the persistent phosphors listed in the tables of this review still have to show their real potential to shine.
While Eu$^{2+}$-doped persistent phosphors are still unrivalled for blue and green emission, the use of other dopants allows one to extend the wavelength range that can be covered with persistent luminescence. Probably, the potential applications, especially in the red and near-infrared range, will be a driving force into further research and developments of new non-Eu$^{2+}$-based materials.

In order to be able to compare experimental research obtained by different research groups on identical or different phosphor compositions, there is an urgent need for a more standardized way of measuring and defining persistent phosphor properties. The standard for measuring light output in $\text{cd/m}^2$ is questionable, since the eye sensitivity shifts to shorter wavelengths at lower light levels [196,262,263]. In addition, the standard way of exciting persistent phosphors, using an unfiltered Xe-arc, containing large amounts of short wavelength ultraviolet radiation, is not a realistic approach and cannot be compared to solar or artificial indoor illumination. Finally, a new standard is needed for quantifying the performance of ultraviolet or infrared emitters: since the eye sensitivity is zero at these wavelengths, photopic units cannot be used and performance should be quoted in radiometric units or numbers of photons.

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