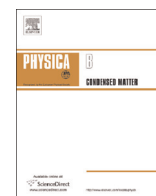




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# Elastico-mechanoluminescence and crystal-structure relationships in persistent luminescent materials and II–VI semiconductor phosphors

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## ABSTRACT

Elastico-mechanoluminescence (EML) has recently attracted the attention of a large number of researchers because of its potential in different types of mechano-optical devices. For understanding the mechanism of EML the relationships between elasto-mechanoluminescence (EML) and crystal-structure of a large number of persistent luminescent materials and II–VI semiconductor phosphors known to date are investigated. It is found that, although most of the non-centrosymmetric crystals exhibit EML, certain non-centrosymmetric crystals do not show EML. Whereas, many centrosymmetric crystals do not exhibit EML, certain centrosymmetric crystals exhibit EML. Piezoelectric ZnS:Cu,Cl single crystals do not show EML, but piezoelectric ZnS:Cu,Cl microcrystalline phosphors show very intense EML. Piezoelectric single crystals of undoped ZnS do not show EML. It seems that EML is related to local piezoelectrification near the impurities in crystals where piezoelectric constant is high. Suitable piezoelectric field near the local piezoelectric region and stable charge carriers in traps are required for appearance of EML. The EML of persistent luminescent materials and II–VI semiconductor phosphors can be understood on the basis of piezoelectrically-induced trap-depth reduction model of EML. Using suitable dopants both in non-centrosymmetric and centrosymmetric crystals intense elasto-mechanoluminescent materials emitting desired colours can be tailored, which may find applications in several mechano-optical devices.

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## 1. Introduction

Recently, elasto-mechanoluminescence (EML), the luminescence induced by elastic deformation of solids, has attracted the attention of a large number of researchers because of its potential in different types of mechano-optical devices such as stress sensors, impact sensor, fracture sensor, torsion sensor, friction sensor, damage sensor, real-time visualization of stress distribution in solids, real-time visualization of stress distribution near crack-tip, real-time visualization of quasidynamic crack-propagation in solids, mechanoluminescence (ML)-based health monitoring system of structures, novel ML-driven photocell system, ML light sources, ML displays, visualization of internal defects in a pipe, stress imaging of the orthopaedic prosthetic devices such as the artificial legs, determination of ultrasonic power, radiation dosimetry, sensing the formation and propagation of Lüders bands, visualization of Portevin–Le Chatelier effect, etc. [1–10]. Jeong et al. [8–

10] have demonstrated that the composite film containing ZnS:Cu, Mn and ZnS:Cu phosphors in a polydimethylsiloxane (PDMS) matrix can be used to produce light source, multicolour displays, and wind-driven light sources.

The elasto-mechanoluminescent materials present an accurate linearity of EML intensity against pressure in the elastic region and they offer the advantages of repetitive, wireless, non-destructive, reproducible, real-time and reliable stress sensing. The processes responsible for the EML excitation in noncentrosymmetric and centrosymmetric crystals are not satisfactorily known till now, and therefore, it is difficult to tailor the intense elasto-mechanoluminescent materials emitting desired colours. The persistent luminescent materials and II–VI semiconductor phosphors have been reported to be prominent elasto-mechanoluminescent materials. The present paper explores the elasto-mechanoluminescence and crystal-structure relationships in persistent luminescent materials and II–VI semiconductor phosphors known to date and shows that most of the noncentrosymmetric crystals and many centrosymmetric crystals exhibit EML. It is shown that the local piezoelectric field near the defects in crystals is responsible for the EML emission in the crystals. It is to be noted

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that if bulk piezoelectricity is responsible for the EML excitation, then all the piezoelectric crystals should exhibit EML and all the non-piezoelectric crystals should not exhibit EML. Such fact is not supported by the experimental results. It is shown that, using suitable dopants both in non-centrosymmetric and centrosymmetric crystals, intense elasto-mechanoluminescent materials emitting desired colours can be tailored, which may find important applications in sensing, imaging, and in many other mechano-optical devices.

## 2. Elastico ML and crystal-structure relationships in persistent

**Table 1**

Correlation between crystal-structure and elasto-mechanoluminescence of crystals.

S. no.	Crystal	Crystal system, space group [references]	Piezoelectric: + non-piezoelectricity: – [references]	ML present: + Absent: – [references]
1	$\alpha$ -SrAl <sub>2</sub> O <sub>4</sub> :Eu	Monoclinic P2 <sub>1</sub>	+ [11]	+ [11]
2	$\beta$ -SrAl <sub>2</sub> O <sub>4</sub> :Eu	Hexagonal P6 <sub>3</sub> 22	+ [11]	– [11]
3	SrAl <sub>12</sub> O <sub>19</sub> :Eu	Hexagonal P6 <sub>3</sub> /mmc	– [11]	– [11]
4	SrAl <sub>4</sub> O <sub>7</sub> :Eu	Hexagonal P6 <sub>3</sub> /mmc	– [11]	– [11]
5	Sr <sub>4</sub> Al <sub>14</sub> O <sub>25</sub> :Eu	Orthorhombic Pmma	– [11]	– [11]
6	Sr <sub>3</sub> Al <sub>2</sub> O <sub>6</sub> :Eu	Cubic Pa3	– [11]	– [11]
7	SrAl <sub>2</sub> O <sub>4</sub> :Eu,Dy	Monoclinic P2 <sub>1</sub> [11,12]	+ [11,12]	+ [3]
8	SrAl <sub>2</sub> O <sub>4</sub> :Ce	Monoclinic P2 <sub>1</sub> [12]	+ [12]	+ [12]
9	SrAl <sub>2</sub> O <sub>4</sub> :Ce,Ho	Monoclinic P2 <sub>1</sub> [12]	+ [12]	+ [12]
10	CaAl <sub>2</sub> O <sub>4</sub> :Eu	Monoclinic (P21/n)	– [13,14]	+ [15,16]
11	MgAl <sub>2</sub> O <sub>4</sub> :Eu	Cubic (Fd3m)	– [14,17]	+ [15,16]
12	CaZr(PO <sub>4</sub> ) <sub>2</sub> :Eu <sup>2+</sup>	Orthorhombic P2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub> [18]	+ [18]	+ [18]
13	BaSi <sub>2</sub> O <sub>2</sub> N <sub>2</sub> :Eu	Orthorhombic Cmc21 [19]	+ [19]	+ [19]
14	SrSi <sub>2</sub> O <sub>2</sub> N <sub>2</sub> :Eu	Triclinic P1 [20]	+ [20]	+ [19]
15	CaSi <sub>2</sub> O <sub>2</sub> N <sub>2</sub> :Eu	Monoclinic P2 <sub>1</sub> [21]	+ [21]	– [19]
16	CaZnOS:Mn <sup>2+</sup>	Hexagonal P6 <sub>3</sub> mc [22]	+ [22]	+ [22]
17	BaTiO <sub>3</sub> –CaTiO <sub>3</sub>	Tetragonal P4mm (ferroelectric phase at room temperature) [23]	+ [24]	+ [24]
18	SrBaMgSi <sub>2</sub> O <sub>7</sub> :Eu	Tetragonal P4 <sub>2</sub> m [25]	+ [25]	+ [25]
19	Sr <sub>2</sub> MgSi <sub>2</sub> O <sub>7</sub> :Eu	Tetragonal [25] P4 <sub>2</sub> m	+ [25]	+ [25]
20	SrCaMgSi <sub>2</sub> O <sub>7</sub> :Eu,	Tetragonal P4 <sub>2</sub> m [25]	+ [25]	+ [25]
21	SrMgAl <sub>6</sub> O <sub>11</sub> :Eu (composed of SrAl <sub>2</sub> O <sub>4</sub> + SrMgAl <sub>10</sub> O <sub>17</sub> + MgAl <sub>2</sub> O <sub>4</sub> phases [29])	Monoclinic P2 <sub>1</sub> [10,11] Hexagonal + P6 <sub>3</sub> /mmc [27,28] Cubic + Fd3m [28]	+ [26] – [27,28] – [28]	+ [26]
22	Ca <sub>2</sub> MgSi <sub>2</sub> O <sub>7</sub> :Eu	Tetragonal P4 <sub>2</sub> m [29]	+ [29]	+ [29]
23	Ca <sub>2</sub> MgSi <sub>2</sub> O <sub>7</sub> :Eu,Dy	Tetragonal P4 <sub>2</sub> m [29]	+ [29]	+ [29]
24	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> :Eu	Triclinic P1 [30]	+ [30]	+ [30]
25	SrAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> :Eu	Monoclinic, I2/c [30–34]	– [30]	+ [30]
26	Ca <sub>1-x</sub> Sr <sub>x</sub> Al <sub>2</sub> SiO <sub>8</sub> :Eu <sup>2+</sup>	Triclinic P1 [30] x=0.91 Monoclinic, I2/c [30–34] For x > 0.91 to 1	+ [30] – [30]	+ [30] + [30]
27	Ca <sub>2</sub> Al <sub>2</sub> SiO <sub>7</sub> :Ce	Tetragonal P4 <sub>2</sub> m [35]	+ [35]	+ [36]
28	SrMg <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> :Eu	Trigonal P3m <sub>1</sub> [37]	+ [37]	+ [38]
29	CaYAl <sub>3</sub> O <sub>7</sub> :Eu	Tetragonal P4 <sub>2</sub> m [39]	+ [39]	+ [39]
30	Sr <sub>n+1</sub> Sn <sub>n</sub> O <sub>3n+1</sub> :Sm <sup>3+</sup> (n = 1, 2, ∞) mono layer double layers	Orthorhombic Pccn [40] Orthorhombic Amam [41]	– [40] – [41]	+ [42] + [42]
31	ZrO <sub>2</sub> :Ti	Monoclinic P2 <sub>1</sub> /c [43–47] (at room temperature) Cubic(at high temperatures > 2370 °C) Fm3m [43,46,47] Tetragonal at intermediate temperatures (1200–2370 °C), P4 <sub>2</sub> /nmc [43,46,47]	– [43–47] – –	+ [48]
32	ZnS:Mn (Wurtzite)	Hexagonal P6mm [22]	+ [22]	+ [49]
33	ZnS:Cu microcrystalline phosphor (Wurtzite) mixed in polydimethylsiloxane (PDMS) elastomer	Hexagonal P6mm [22]	+ [22]	+ [8–10]
34	ZnS:Mn,Cu (Wurtzite) microcrystalline phosphor (Wurtzite) mixed in PDMS elastomer	Hexagonal P6mm [22]	+ [22]	+ [8–10]
35	CdS:Ag (Wurtzite) (Low temperature)	Hexagonal P6mm [22]	+ [22]	+ [50]
36	ZnMnTe	Hexagonal, P6mm [51] + Zinc blende, Fm3m [51]	+ [51] – [51]	+ [51]
37	(ZnS) <sub>1-x</sub> (MnTe) <sub>x</sub>	Hexagonal (Wurtzite) P6mm [52]	+ [52]	+ [52]
38	(ZnS) <sub>1-x</sub> (MnTe) <sub>x</sub>	Cubic (sphalerite) F43m [53]	+ [53]	+ [53]
39	ZnS:Cu,Cl (single crystals)	Hexagonal (Wurtzite) P6mm [22]	+ [22]	– [54]
40	ZnS (undoped single crystals)	Hexagonal (Wurtzite) P6mm [22]	+ [22]	– (Our observation)

## luminescent materials and II–VI semiconductor phosphors

Table 1 shows the correlation between the elasto ML and crystal-structure of a large number of persistent luminescent materials and II–VI semiconductor phosphors known to date, in which the space groups and occurrence of EML of the crystals were taken from the references [11–54]. The following inferences can be drawn from the results shown in Table 1.

- (1) Except a few materials such as  $\beta$ -SrAl<sub>2</sub>O<sub>4</sub>, CaSi<sub>2</sub>O<sub>2</sub>N<sub>2</sub>:Eu, ZnS:Cu,Cl single crystals and undoped ZnS single crystals, all the other piezoelectric (non-centrosymmetric) crystals shown in Table 1 exhibit EML.
- (2) Some of the centrosymmetric crystals such as CaAl<sub>2</sub>O<sub>4</sub>:Eu,

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