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α-Sr2Si2O2N2 is one of the recently studied oxonitridosilicates applicable in optoelectronics, in particular in white LEDs. Its elastic properties remain unknown. A survey of literature shows that, up to now, nine oxonitridosilicate materials have been identified. For most of these compounds, doped with rare earths and manganese, a luminescence has been reported at a wavelength characteristic for the given material; all together cover a broad spectral range. The present study focuses on the elastic properties of one of these oxonitridosilicates, the Eu-doped triclinic α-SrSi2O2N2. High-pressure powder diffraction experiments are used in order to experimentally determine, for the first time, the equation of state of this compound. The in situ experiment was performed for pressures ranging up to 9.65 GPa, for Eu-doped α-SrSi2O2N2 sample mounted in a diamond anvil cell ascertaining the hydrostatic compression conditions. The obtained experimental variation of volume of the triclinic unit cell of α-SrSi2O2N2:Eu with rising pressure served for determination of the Birch-Murnaghan equation of state. The determined above quoted bulk modulus is 103(5) GPa, its first derivative is 4.5(1.1). The above quoted bulk modulus value is found to be comparable to that of earlier reported oxynitrides of different composition. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4883502]

I. INTRODUCTION

Oxonitrides form a large class of crystalline and amorphous materials having various potential applications.1,2 Physicochemical properties of oxynitrides differ from those of oxides, due to the presence of the cation-nitrogen bonds which are more covalent than the cation-oxygen bonds. Oxynitrides exhibit, in particular, valuable luminescence properties. Their applications as phosphors have been reviewed in Refs. 3–6.

An important subclass of oxynitrides is formed by oxonitridosilicates.7 Among them, the compounds belonging to the Sr-Si-O-N system recently have attracted attention of a number of researchers developing new optoelectronic materials. In this system, nine quaternary materials differing by composition or structure have already been synthesized (see Table I): two polymorphs of Sr2Si5N8−xOx,19−21 monoclinic Sr3Si2O4N2,10,11 Sr3Si6O12N2,12 triclinic SrSi2O2N2:Eu with rising pressure served for determination of the Birch-Murnaghan equation of state. The determined above quoted bulk modulus is 103(5) GPa, its first derivative is 4.5(1.1). The above quoted bulk modulus value is found to be comparable to that of earlier reported oxynitrides of different composition. © 2014 AIP Publishing LLC.

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TABLE I. Quaternary oxonitridosilicates in the Sr-Si-O-N system, the list is completed by two Ba-containing solid solutions. The numbers in the first column distinguish structurally different compounds. The optoelectronic applications listed concern doped samples using dopants named in the third column. NR—Not reported.

<table>
<thead>
<tr>
<th>No.</th>
<th>Strontium oxonitridosilicate</th>
<th>Dopant</th>
<th>Si/(Si + Sr)</th>
<th>N/(N + O)</th>
<th>Crystal system</th>
<th>Space group</th>
<th>Prospective applications of doped material</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Sr₂SiO₅₋₁S₅N₈₋₄</td>
<td>Eu</td>
<td>0.33</td>
<td>Varied</td>
<td>Monoclinic</td>
<td>P12₁/n1</td>
<td>Phosphor (red) for white LEDs</td>
<td>8</td>
</tr>
<tr>
<td>2</td>
<td>Sr₂SiO₅₋₁S₅N₈₋₄</td>
<td>Eu</td>
<td>0.33</td>
<td>Varied from 0.192 to 0.352 (0.7 &lt; x &lt; 1.2)</td>
<td>Orthorhombic</td>
<td>Pnma</td>
<td>Phosphor (red)</td>
<td>9</td>
</tr>
<tr>
<td>3</td>
<td>Sr₃Si₂O₄N₂</td>
<td>Eu, Ce, Li</td>
<td>0.40</td>
<td>0.33</td>
<td>Cubic</td>
<td>Pn̅3</td>
<td>Phosphor (blue-UV) for white LEDs</td>
<td>10, 11</td>
</tr>
<tr>
<td>4</td>
<td>Sr₃Si₂O₄N₂</td>
<td>Eu</td>
<td>0.67</td>
<td>0.14</td>
<td>Hexagonal</td>
<td>P-3</td>
<td>Phosphor (green), white LEDs</td>
<td>12</td>
</tr>
<tr>
<td>5</td>
<td>Sr₂SiO₂N₂</td>
<td>Eu</td>
<td>0.67</td>
<td>0.50</td>
<td>Triclinic</td>
<td>P1</td>
<td>Phosphor (yellow/red) for white LEDs</td>
<td>13–15</td>
</tr>
<tr>
<td>6</td>
<td>Sr₃Si₂O₄N₂</td>
<td>Eu</td>
<td>0.67</td>
<td>0.33</td>
<td>Orthorhombic</td>
<td>Pnma</td>
<td>Phosphor (blue-UV) for white LEDs</td>
<td>16</td>
</tr>
<tr>
<td>7</td>
<td>Sr₂SiO₂N₂</td>
<td>Eu</td>
<td>0.67</td>
<td>0.73</td>
<td>Monoclinic</td>
<td>P21</td>
<td>White LEDs</td>
<td>17</td>
</tr>
<tr>
<td>8</td>
<td>Sr₃Si₂O₄N₂</td>
<td>Eu,Mn</td>
<td>0.71</td>
<td>Varied</td>
<td>Orthorhombic</td>
<td>Pnma₂₁</td>
<td>Phosphor (blue-UV)</td>
<td>19</td>
</tr>
<tr>
<td>9</td>
<td>Sr-Si-O-N glass</td>
<td>…</td>
<td>0.60 (middle of the 0.45 to 0.75 range)</td>
<td>Amorphous</td>
<td>…</td>
<td>Refractive glass</td>
<td>21, 22</td>
<td></td>
</tr>
</tbody>
</table>

*The Sr₃SiN₈ compound has been reported to be a red phosphor.†*

As a consequence, SrSi₂O₂N₂:Eu²⁺ can be used in green LEDs, and as a green component in a two- or three-component phosphor mixture, that enable to achieve a desired high Color Rendering Index (CRI) of a white LED. An application in commercial LEDs has been mentioned in Ref. 32. Co-doping with Eu and Mn has been recently reported to provide the green color alternating current LEDs. The luminescence of SrSi₂O₂N₂:Eu²⁺ can be additionally enhanced by co-doping with Ce³⁺, Dy³⁺, Mn²⁺, and Mg²⁺ ions. In spite of the fact that SrSi₂O₂N₂:Eu²⁺ is an excellent conversion phosphor material for LEDs, it also exhibits persistent luminescence. Recently, Tang et al. have reported a successful preparation of core–shell structured SiO₂@SrSi₂O₂N₂ phosphor: the particles with outer h-BN film, the inner SrSi₂O₂N₂:Eu²⁺ shell, and amorphous SiO₂ cores exhibit similar spectral features to those of bulk SrSi₂O₂N₂:Eu²⁺ prepared by solid state reaction.

Structures of SrSi₂O₂N₂ polymorphs have been studied only recently, whereas the structural and elastic properties under non-ambient pressure or temperature remain unknown.

According to a study employing a combined X-ray diffraction and electron diffraction, SrSi₂O₂N₂ adopts a triclinic structure (space group P1), hereafter referred to as the α phase. In the cited paper, an approximate structure model has been proposed, being derived from the structure of EuSi₂O₂N₂ determined in Ref. 39. In the proposed SrSi₂O₂N₂ unit cell, the Sr atoms partially (∼20%) occupy new lattice sites, moreover, the presence of spurious phases, the occurrence of twinning and domain intergrowth lead to difficulties in structure refinement. The chemically related BaSi₂O₂N₂ and CaSi₂O₂N₂ exhibit higher symmetry orthorhombic Pbcn and monoclinic P2₁ structures, respectively. Recently, it has been shown that SrSi₂O₂N₂ can crystallize as a previously unknown monoclinic phase (hereafter referred to as a β phase). Its structure, of a marginally smaller density than that of the α phase, was proposed in Ref. 18, but detailed structural data on the β phase are not yet available.

The α-SrSi₂O₂N₂:Eu²⁺ is usually prepared by a solid state synthesis method. It is difficult to obtain a pure-phase material: the synthesis procedure may influence the molar ratio of the α and β phases in the product (e.g., the β phase has been shown to appear when Sr₂SiO₄ was used as a precursor). Reported structural data for α and β SrSi₂O₂N₂ phases are quoted in Table II.

Equation of state describes fundamental physicochemical properties of a substance. Its coefficients (bulk modulus and its derivatives) are dependent on the nature and strength of the interatomic bonds. The knowledge of the elastic properties (lattice parameters variation, bulk modulus value) of a material allows for understanding of its behavior under external pressure and also under so called “chemical” pressure induced by, for example, doping with ions of different ionic...
TABLE II. Reported lattice parameters for undoped and rare-earth-doped SrSi2O2N2 and for EuSi2O2N2. Phase content of the given phase in samples containing both α and β phases is provided in the third column; the values of phase content given in parentheses are roughly evaluated here on the basis of peak heights. nq: Not quoted.

<table>
<thead>
<tr>
<th>Formula</th>
<th>Space group</th>
<th>Phase content [%]</th>
<th>(a) [Å]</th>
<th>(b) [Å]</th>
<th>(c) [Å]</th>
<th>(\alpha)</th>
<th>(\beta)</th>
<th>(\gamma)</th>
<th>(V) [Å³]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Undoped (\alpha) phase</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\alpha)-SrSi2O2N2</td>
<td>(P1)</td>
<td>&gt;93(^a)</td>
<td>7.0802(2)</td>
<td>7.2306(2)</td>
<td>7.2554(2)</td>
<td>88.767(3)</td>
<td>84.733(2)</td>
<td>75.905(2)</td>
<td>358.73(2)</td>
<td>15</td>
</tr>
<tr>
<td>(\alpha)-SrSi2O2N2</td>
<td>(P1)</td>
<td>&gt;95</td>
<td>7.1014(4)</td>
<td>7.2522(4)</td>
<td>7.2785(8)</td>
<td>88.771(1)</td>
<td>84.754(4)</td>
<td>75.919(1)</td>
<td>362.02(8)</td>
<td>46</td>
</tr>
<tr>
<td>EuSi2O2N2</td>
<td>(P1)</td>
<td>...</td>
<td>7.0950(10)</td>
<td>7.2460(10)</td>
<td>7.2560(10)</td>
<td>88.69(2)</td>
<td>84.77(2)</td>
<td>75.84(2)</td>
<td>360.19</td>
<td></td>
</tr>
<tr>
<td>RE doped (\alpha) phase</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\alpha)-Sr0.95Th0.05Si2O2N2</td>
<td>(P1)</td>
<td>...</td>
<td>7.1511</td>
<td>7.3021</td>
<td>7.3337</td>
<td>88.88</td>
<td>84.78</td>
<td>76.04</td>
<td>370.10</td>
<td>43</td>
</tr>
<tr>
<td>(\alpha)-Sr0.95Ce0.05Si2O2N2</td>
<td>(P1)</td>
<td>...</td>
<td>7.1574</td>
<td>7.3071</td>
<td>7.3343</td>
<td>88.81</td>
<td>84.74</td>
<td>76.08</td>
<td>370.75</td>
<td>43</td>
</tr>
<tr>
<td>(\alpha)-Sr0.95Ba0.05Si2O2N2</td>
<td>(P1)</td>
<td>...(^b)</td>
<td>7.2059(2)</td>
<td>7.3887(3)</td>
<td>7.3430(2)</td>
<td>88.525(4)</td>
<td>84.454(3)</td>
<td>75.797(4)</td>
<td>377.07</td>
<td>44</td>
</tr>
<tr>
<td>(\alpha)-Sr0.9Eu0.04Si2O2N2</td>
<td>(P1)</td>
<td>&gt;80</td>
<td>7.1106(7)</td>
<td>7.2625(0)</td>
<td>7.2899(7)</td>
<td>nq</td>
<td>nq</td>
<td>nq</td>
<td>363.48(2)</td>
<td>33</td>
</tr>
<tr>
<td>(\alpha)-Sr0.9Eu0.07Si2O2N2</td>
<td>(P1)</td>
<td>nq</td>
<td>nq</td>
<td>nq</td>
<td>nq</td>
<td>nq</td>
<td>nq</td>
<td>nq</td>
<td>361.95</td>
<td></td>
</tr>
<tr>
<td>(\alpha)-SrSi2O2N2:Eu</td>
<td>(P1)</td>
<td>98</td>
<td>7.1025(2)</td>
<td>7.2535(2)</td>
<td>7.2813(2)</td>
<td>88.732(3)</td>
<td>84.768(2)</td>
<td>75.886(3)</td>
<td>362.28(2)</td>
<td></td>
</tr>
</tbody>
</table>

| RE doped \(\beta\) phase |
| \(\beta\)-SrSi2O2N2:Eu | \(P2_1\) | 48                | 7.1036(14)| 14.078(3) | 7.2833(15)| 90        | 95.23(3)  | 90        | 725.3(3)  | 18        |

\(^a\)Impurity phases Sr2N2O, SrSiO2, and amorphous components detected.
\(^b\)Details given in the reference.

radii or by stress induced in the layered structures on the boundaries between various layers of different lattice parameters. It is of great importance since the luminescence in these materials doped with rare-earth ions is often associated with the 5d-4f optical transition, as in the case of Eu\(^{2+}\). Thus, the transition energy is strongly dependent on the strength of the crystal field, i.e., on the distance \(r\) between the central ions and ligands (it is scaled approximately as \(r^{-5}\)). Such structural engineering leads to opportunity of tuning of the photoluminescence, creating a way to achievement of a desired wavelength and color rendering index.\(^{46}\)

The present paper reports the determination of the yet unknown equation of state (EOS) of \(\alpha\)-SrSi2O2N2. The results, based on X-ray diffraction study under hydrostatic pressure, show that in the pressure range studied (up to 9.65 GPa) its triclinic structure is conserved and provides the basic information about the elastic properties of this material.

II. EXPERIMENTAL DETAILS

The studied SrSi2O2N2:Eu\(^{2+}\) sample, with intentional Eu dopant concentration of 2%, was prepared by high-temperature solid state reaction method. The starting materials were high purity SrCO3, SiO2, Si3N4, and Eu2O3. To obtain a stoichiometric homogeneous mixture, the appropriate amounts of the reagents were thoroughly ground. The powder was sintered at 1400–1600 °C for 1 h under reducing atmosphere of H2 (25%–N2 (75%). After sintering, the sample was cooled down to room temperature and then reground (details of the procedure follow those of Ref. 34). The obtained sample was of white color with a light greenish shade.

Phase analysis and structure refinement were performed on the basis of laboratory powder diffraction data. The measurement was conducted at \(T = 297(3)\) K using Philips X’Pert Pro MPD Bragg-Brentano diffractometer equipped with an incident Ge(111) monochromator and a strip detector. The experiment was performed using Cu \(K\alpha_1\) radiation.

The \(\text{in situ}\) high-pressure diffraction was conducted at the I711 beamline of MAX II storage ring (MAX IV Laboratory, Lund, Sweden) at room temperature, using a diamond anvil cell. The measurements were carried out under hydrostatic conditions (methanol-ethanol-water mixture 16:3:1 as a pressure-transmitting medium) with a monochromatic radiation of 0.9917 Å wavelength. The diffracted beam was registered by an area detector (Titan CCD, Agilent), which was placed at a distance of 8.80 cm from the specimen. A preindented gasket of 100 μm thickness with a centered hole of 200 μm diameter was used in the experiment: the powdered sample and a ruby chip serving as a pressure gauge, both immersed in the pressure transmitting medium filling the hole were placed between the diamond anvils. The powder diffraction patterns covered the 2θ angles ranging up to 35°. For structure refinement, the Rietveld refinements using the Fullprof.2k software\(^{47}\) were performed. The 2D diffraction patterns collected with the area detector were converted to 1D patterns with the Fit2D software.\(^{48}\) The Birch-Murnaghan equation of state was determined from the experimental data on lattice parameter and volume variations on pressure, using the EOSfit52 program.\(^{49}\) The calculation of the elasticity tensor was performed using the STRAIN program.\(^{50}\)

III. RESULTS

A. Phase analysis and Rietveld refinement

Phase analysis shows that the main phase of the sample is the \(\alpha\)-SrSi2O2N2 (space group \(P1\), \(Z = 4\)) with a trace amount of the \(\beta\) polymorph. The presence of the latter is documented by the characteristic diffraction peaks at angles 14.01° (\(hkl\) = 110, intensity of 0.3%), 17.75° (120/−111, 0.2%), 22.31° (130, 22.7 (0.2%), 0.5%), 26.60° (−102, 3%), 27.3 (−112, 1.2%), 28.06 (102, 3%), 29.35° (112, 0.7%), 29.50° (−122, 0.3%) (in agreement with Ref. 18); other lines are not detectable due to their low intensity and a strong overlap with...
those of the $\alpha$ phase. Judging after the intensity of the $-102$ reflection, the amount of $\beta$ phase is here distinctly smaller than in the sample studied in Ref. 15. Based on a simple comparison of peak intensities relationships with those of the data of two-phase sample of Ref. 18, the content of the $\beta$ phase is roughly evaluated as about 2%. The Rietveld refinement was performed using a similar approach as that in Ref. 15 the oxygen and nitrogen positions were not refined, and two very small angular ranges where the most intense spurious peaks appear were excluded. The lattice parameters of the $\alpha$ phase at temperature of 297(3) K are $a = 7.1025(2)$ Å, $b = 7.2535(2)$ Å, $c = 7.2813(2)$ Å, $\alpha = 88.732(3)$°, $\beta = 84.768(2)$°, $\gamma = 75.886(3)$°, $V = 362.28(2)$ Å$^3$ and the calculated density is 3.737 g/cm$^3$.

No systematic studies of $\alpha$-SrSi$_2$O$_2$N$_2$ unit-cell size variation with doping could be found in literature, except that for Ba substitution at Sr sites (a monotonic increase is reported$^{45}$). Substituting a small amount of Eu$^{2+}$ ions at Sr sites is known to have no significant influence on the cell volume; all six lattice parameters differ marginally between the side compounds, as Table II shows. Figure 1 displays the unit cell volume versus the content of the substituting rare-earth element. The dashed line shows, based on points reported by different groups (Refs. 15, 17, 33, 45, 39, and this work), that the volume does not vary with Eu substitution. The scatter of the experimental points (especially that for Ce and Yb points added to the figure) can be attributed to differences in technology leading to compositional differences (such as a deviation in N/(N+O) ratio).

B. Equation of state

For the determination of equation-of-state parameters, diffraction data for 13 pressure points were collected at pressures ranging up to 9.65 GPa. When the pressure increases in the range up to 9.65 GPa, the structure of the SrSi$_2$O$_2$N$_2$ sample is conserved, as documented by the diffraction patterns (Fig. 2). The starting point in the Rietveld refinement of compressed sample was based on the $\alpha$ phase structure$^{15}$. As the amount of the $\beta$ phase was marginal, the reflections of this phase could not be observed in the high-pressure experiment, so this component was not included in the refinements. The refinements provided the lattice parameters which together with unit-cell volumes and densities are quoted in Table III. The $a$, $b$, and $c$ values decrease smoothly as a function of applied pressure in the whole studied pressure range (see Fig. 3).

To determine the parameters of EOS for the investigated sample, the third-order Birch-Murnaghan (BM) equation of state (1)$^{51}$ was used as

$$ p = \frac{3}{2} K_0 \left\{ \left( \frac{V_0}{V} \right)^{\frac{3}{2}} - \left( \frac{V_0}{V} \right)^{\frac{7}{2}} \right\} \left\{ 1 + \frac{3}{4} \left[ K'_0 - 4 \right] \right\} \times \left[ \left( \frac{V_0}{V} \right)^{\frac{3}{2}} - 1 \right], $$

where $p$ represents the pressure, $V$ is the volume, $K$ is the isothermal bulk modulus defined as $K = -V(\partial p/\partial V)$, $K'$ is the first derivative of bulk modulus: $K' = -V(\partial K/\partial p)$, and $V_0$, $K_0$, $K'_0$ refer to volume, bulk modulus, and derivative of bulk modulus, respectively, at $p = 0$. The fitting was performed...
TABLE III. Variation of the SrSi$_2$O$_2$N$_2$:Eu unit cell size and material density with rising pressure.

<table>
<thead>
<tr>
<th>p (GPa)</th>
<th>a (Å)</th>
<th>b (Å)</th>
<th>c (Å)</th>
<th>α (deg)</th>
<th>β (deg)</th>
<th>γ (deg)</th>
<th>V (Å$^3$)</th>
<th>Density (g/cm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.96</td>
<td>7.080(1)</td>
<td>7.235(2)</td>
<td>7.255(1)</td>
<td>88.99(2)</td>
<td>84.82(1)</td>
<td>75.83(3)</td>
<td>358.9(2)</td>
<td>3.772</td>
</tr>
<tr>
<td>1.45</td>
<td>7.072(1)</td>
<td>7.222(2)</td>
<td>7.244(1)</td>
<td>89.02(2)</td>
<td>84.83(1)</td>
<td>75.82(3)</td>
<td>357.8(2)</td>
<td>3.789</td>
</tr>
<tr>
<td>1.95</td>
<td>7.064(1)</td>
<td>7.211(2)</td>
<td>7.229(1)</td>
<td>89.07(2)</td>
<td>84.86(1)</td>
<td>75.77(3)</td>
<td>355.9(2)</td>
<td>3.808</td>
</tr>
<tr>
<td>2.63</td>
<td>7.0539(9)</td>
<td>7.192(2)</td>
<td>7.218(1)</td>
<td>89.02(2)</td>
<td>84.89(1)</td>
<td>75.76(3)</td>
<td>353.5(2)</td>
<td>3.829</td>
</tr>
<tr>
<td>3.23</td>
<td>7.045(1)</td>
<td>7.178(2)</td>
<td>7.203(1)</td>
<td>89.04(2)</td>
<td>84.90(1)</td>
<td>75.77(3)</td>
<td>351.8(2)</td>
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</tr>
<tr>
<td>3.92</td>
<td>7.0375(9)</td>
<td>7.157(2)</td>
<td>7.187(1)</td>
<td>89.14(2)</td>
<td>84.99(1)</td>
<td>75.87(3)</td>
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<tr>
<td>4.60</td>
<td>7.0223(9)</td>
<td>7.140(2)</td>
<td>7.173(1)</td>
<td>89.08(2)</td>
<td>84.98(1)</td>
<td>75.87(3)</td>
<td>347.3(2)</td>
<td>3.897</td>
</tr>
<tr>
<td>5.54</td>
<td>7.012(1)</td>
<td>7.126(2)</td>
<td>7.158(1)</td>
<td>89.08(2)</td>
<td>85.00(1)</td>
<td>75.78(3)</td>
<td>345.3(2)</td>
<td>3.919</td>
</tr>
<tr>
<td>6.36</td>
<td>7.001(1)</td>
<td>7.103(2)</td>
<td>7.140(1)</td>
<td>89.11(2)</td>
<td>85.06(1)</td>
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<tr>
<td>7.32</td>
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<td>7.122(1)</td>
<td>89.09(2)</td>
<td>85.08(1)</td>
<td>75.69(3)</td>
<td>342.0(2)</td>
<td>3.978</td>
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<tr>
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<td>7.107(1)</td>
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<td>3.999</td>
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<td>7.083(1)</td>
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<td>85.22(2)</td>
<td>75.70(3)</td>
<td>335.0(3)</td>
<td>4.040</td>
</tr>
</tbody>
</table>

according to the strategy described in Ref. 52; for modeling of the lattice parameter variation, closely related equations described in the cited reference were used.

The unit-cell angles do not vary much: $\alpha$ and $\gamma$ are almost constant whereas the value of $\beta$ angle slightly increases with rising pressure (see Fig. 4). The difference between the compressibility in the $a$, $b$, and $c$ directions is manifested by different slopes of the curves in Fig. 3. The compressibility along the axes is not identical: the material is most compressible in the $b$ direction (3.2% of axis length reduction in the studied range) whereas it is less compressible in the $a$ and $c$ directions (1.8% and 2.7%, respectively). In order to quantitatively determine the compressibility along the crystallographic axes, the pressure variations of $a$, $b$, and $c$ were fitted with the third-order Birch-Murnaghan EOS. The procedure provided the following parameters: $a_0 = 7.094(1)$ Å, $K_{0a} = 145(5)$ GPa, $K'_{0a} = 4.5(1.5)$, $b_0 = 7.264(1)$ Å, $K_{0b} = 84(2)$ GPa, $K'_{0b} = 3.6(6)$, $c_0 = 7.277(1)$ Å, $K_{0c} = 99(2)$ GPa, $K'_{0c} = 4.0(6)$. Elasticity tensor calculation shows that direction of the softest compressibility is close to the [010] direction. It may be expected that the direction of the strongest thermal expansion is the same.

It is noteworthy that despite the differences in the laboratory data collection method and obtained statistics, the experimental points for lattice parameters, angles and volume are located almost ideally at the extrapolated lines determined at varying pressures. This behavior confirms the good quality of the applied data analysis procedures.

The BM equation for Eu-doped SrSi$_2$O$_2$N$_2$ was fitted based on the experimental unit cell volume changing with pressure (cf. Fig. 5). Zero-pressure bulk modulus $K_0$ for SrSi$_2$O$_2$N$_2$:Eu was calculated to be 103(5) GPa with a first derivative $K'_0$ of 4.5(1.1), the implied second derivative $K'' = -0.04558$ GPa$^{-1}$, and fitted zero-pressure volume $V_0 = 362.1(3)$ Å$^3$. The increase of the bulk modulus with pressure, derived from the obtained EOS, is shown in Fig. 6.

![FIG. 3. Unit cell parameters $a$, $b$, and $c$ of $\alpha$-SrSi$_2$O$_2$N$_2$:Eu (●) as a function of pressure. Ambient pressure values obtained in the laboratory conditions (♦). The lines correspond to Birch-Murnaghan lattice parameter fit described in the text. Standard deviations are smaller than the symbol size.](image)

![FIG. 4. Unit-cell angles of $\alpha$-SrSi$_2$O$_2$N$_2$:Eu as a function of pressure (●). Ambient pressure values obtained in the laboratory conditions (♦). The lines are a guide to the eye. Standard deviations are smaller than the symbol size.](image)
Reported data on bulk modulus for compounds in the Sr-Si-O-N system for binary\textsuperscript{53–56} and ternary\textsuperscript{57} compounds, for a quaternary compound (the present result), as well as for some other oxynitrides\textsuperscript{58–62}, oxycarbides\textsuperscript{63}, and complex nitrides\textsuperscript{64} are quoted in Table IV. As demonstrated in Table IV(A), the binary nitrides show much higher bulk modulus value than the oxides do. This is due to more covalent character of the bonding in nitrides. Consequently, the bulk modulus in oxynitrides is intermediate, and tends to increase with rising N/(N+O) ratio, as illustrated by the values of 103 GPa for SrSi\textsubscript{2}O\textsubscript{2}N\textsubscript{2}:Eu (the present result) and 130 GPa for Si\textsubscript{2}N\textsubscript{2}O characterized by a higher value of the ratio. Among various oxynitrides, the present value is a moderate one (cf. Table IV(C)). Interestingly, the present value is not much smaller from those for some other oxonitridosilicates such as SrSiAl\textsubscript{2}O\textsubscript{4}N\textsubscript{2} and Ce\textsubscript{2}[Si\textsubscript{4}O\textsubscript{4}N\textsubscript{6}]O, but much smaller than those for nitrogen-rich oxycarbides and nitrides listed in Table IV. It is worth noting that a related effect has been observed for glasses, namely a nitridation of silicate glasses leads to increase of their bulk modulus, as demonstrated in Ref.\textsuperscript{57} and mentioned in Ref.\textsuperscript{65}. On the basis of all above-considered observations, one should expect that among the oxonitridosilicates listed in Table I, those with the highest N/(N+O) ratio will exhibit the largest bulk modulus value and vice versa.

### IV. SUMMARY

Multicomponent oxynitrides form a class of valuable materials applicable as phosphors emitting light of specific colors or as components of phosphor-converted white LEDs. Their elastic properties are not yet known in detail. In particular, the equation of state has not been reported for any of strontium oxonitridosilicates. The present study provides the first experimental data about bulk modulus and its derivative of one of such materials, the $\alpha$-SrSi\textsubscript{2}O\textsubscript{2}N\textsubscript{2}:Eu oxonitridosilicate. The experiments were performed using the \textit{in situ} high-pressure powder diffraction technique under hydrostatic conditions in the pressure range up to 9.65 GPa. The bulk modulus, its derivative, and the unit-cell volume at zero pressure were determined by fitting the third-order Birch-Murnaghan equation of state. The value of bulk modulus is 103(5) GPa with pressure derivative of 4.5(1.1), the zero-pressure
volume is 362.1(3) Å³. The obtained value of bulk modulus is found to be lower than those of two other multinary oxynitrides known from literature. Among strontium oxinitridosilicates, high values of bulk modulus are expected among those of high nitrogen to oxygen ratio.

ACKNOWLEDGMENTS

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60 A. Friedrich, K. Knorr, A. Lieb, S. Rath, M. Hanfland, B. Winkler, and W. Schnick, “High-pressure phase transition of the oxonitridosilicate chloride Ce$_{2}$(Si$_3$O$_{7-x}$N$_{x}$)$_{2}$Cl$_{1-x}$O$_{x}$ with x = 0.12 and 0.18,” Z. Kristallogr. 220(2–3), 245–249 (2005).