Intense piezoluminescence in LiTaO₃ phosphors doped with Pr³⁺ ions

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ABSTRACT

A novel piezoluminescent (PZL) phosphor was prepared by doping Pr³⁺ ions into ferroelectric LiTaO₃ matrices, and their crystal structure and multi-luminescent performance were investigated. A slightly Li-rich non-stoichiometric design gave the phosphors optimized PZL behavior with favorable recoverability. Excited by 240 nm radiation, two strong photoluminescent (PL) emission bands lay around 511 and 615 nm, which corresponded to Pr³⁺ transitions of ⁴P₀ to ⁴H₄ and ¹D₂ to ⁴H₄, respectively. Nevertheless, the ³P₀ to ³H₄ transition was detectably suppressed in AG/PZL spectra with ¹D₂ to ⁴H₄ becoming dominant, which was possibly owing to the effect of trap level distribution on direct tunneling possibilities. In this context, a probable mechanism was proposed to explain the multi-luminescent process in LiTaO₃:Pr³⁺. This study was aimed at exploring novel PZL materials for fundamental research and applications in multifunctional devices.

1. Introduction

Piezoluminescence (PZL) is a typical subcategory of the mechanoluminescence (ML) phenomenon discovered both in organics and inorganics in which certain materials convert compressive mechanical stimuli into light radiation during nondestructive deformation, which was formally introduced by Atari in 1982 [1]. Phosphors of intense PZL are recognized as prospective media for realizing efficient mechano-optical conversion, and such energy transformation mechanisms have generated worldwide attention due to increasing environmental concerns [2]. Near the end of the 20th century, reports of PZL in the elastic region of deformation, also termed elastico-mechanoluminescence (EML), appeared and a new avenue in the field of ML was explored [3–7]. The ground-breaking demonstration of intense, sensitive and repeatable PZL from SrAl₂O₄:Eu²⁺ and ZnS:Mn²⁺, with their early-stage applications, actuated a renewed enthusiasm among scientists in EML materials and promoted development of this field [5,6]. Compared with destructive ML materials (also called fractoluminescent materials) or plastico-mechanoluminescent (PML) materials, materials with PZL, accomplished by elastic deformation, exhibit prominent advantages including recoverable and proportional dependence of luminescence intensity with stress and loading rate [8,9], which are important prerequisites for smart sensors. Therefore, the present research attention on ML has been primarily paid to such PZLs [10], and PZL is broadly taken to refer to PZL in the elastic region of deformation in this context. To date, broad and successful attempts to realize applications based on PZL, such as real-time stress sensing, personal signature recognition, impact sensor, structural and health-monitoring sensors, stress field visualization, and low-cost kinesiological sensors [11–16], have impelled scientists to exploit novel material systems with intense PZL to enrich the family of ML materials, deeply excavate the intrinsic mechanism, and ultimately enhance their social value.

The gradually revealed mechanism behind PZL has provided clues that have facilitated the discovery of novel PZL materials. Varying from fractoluminescence and triboluminescence, which are considered to derive from plasma discharge in the air surrounding a broken surface [17–19], PZL is speculated to originate from multicopling actions that consist of photoluminescent centers, trap states for the storage of charge carriers (electrons and/or holes), and stress-induced piezoelectric fields in favor of detrapping of charge carriers [20–22]. Considering the complicated but intuitive relationship between piezoelectricity and PZL, the objective piezoelectric materials are promising candidates for hosts that are expected to exhibit PZL with appropriate introduction of luminescent ions. The introduced luminescent ions are selected to simultaneously create emission centers and charge-carrier trap states. In terms of choosing appropriate luminescent centers as activators for fascinating optical performance, trivalent rare earth ions (Ln³⁺) are preferred elements for their native superiority from abundant interior energy levels of 4f configuratons. Therein, the energy levels of Pr³⁺ ions endow Pr³⁺-containing phosphors with various energy transfer potentials, where the main emission bands variously locate in the cyan and/or red regions [23–25]. Attractively, several PZL Pr³⁺-containing...
materials have been discovered based on this strategy, including Ba-
TiO3–CaTiO3:Pr3+ [26], Ca3Nb2O7:Pr3+ [22], LiNbO3:Pr3+ [27] and
Ca3Ti2O7:Pr3+ [21]. Pr3+ ions might be a highly matched PZL dopant
for existing piezoelectric matrices. Also, transition metal elements can
also be adopted to function as the abovementioned luminescent ions
and the representative material systems include ZnS:Me2+/Cu+ [6,28]
and CaZnOS: Mn2+/Cu+ [29,30]. In addition, self-PZL, as an optimal
response to an increasingly serious resource problem, is an emerging
and challenging orientation in which PZL occurs with no need for the
inclusion of external dopants. Currently, an undoped CaZnOS piezo-
electric semiconductor has been reported to show intense PZL, re-
acting to various mechanical stimuli through a proposed lumines-
cent mechanism mediated by intrinsic oxygen vacancies [20]. The
scientific payoffs mentioned above indicate a practicable and promising
strategy for producing piezoluminescent materials through existing
piezoelectrics with multifunctional applications. Interactively, the
intrinsic mechanism behind the PZL phenomenon is expected to be
thoroughly illuminated by deeper study.

There is a mainstream trend to realize the integration of multi-
energy conversions in an individual material [26]. A typical instance is
LiNbO3:Pr3+ ceramics, which exhibit simultaneous realization of pie-
zoelectricity with sensitive PZL, and their multipiezo properties are
ideal for smart devices [27]. LiTaO3, as an isomorphous form of LiNbO3,
possesses similar crystal structure and piezoelectric properties. Ac-
cording to the abovementioned factors, an intense PZL in rare earth
doped LiTaO3 was inferred, which has never been reported to our
knowledge. Lithium tantalate, LiTaO3, has been recognized as a pro-
minent lead-free ferroelectric material with extensive applications in
pyroelectric, piezoelectric, and electrooptic devices owing to its favor-
able comprehensive performance [31–34]. Intense research with ever-
growing interest into LiTaO3 doped with rare earth elements has been
performed to examine its luminescent properties as well as its crystal
structures [35–40]. Whereas, these reports of doped LiTaO3 have been
mainly examined for the growth, properties and applications of single
 crystals [41], which are limited by a long production cycle and high
 costs. In addition, limited studies have focused on LiTaO3 ceramics in
the form of a single phase, while these materials serve as end-members
that are introduced to modify the piezoelectric properties of some ty-
pical lead-free ceramic systems, such as (Bi, Na)TiO3 and (K, Na)NbO3
[42–44]. More trials should be performed to tap the potential in LiTaO3
 ceramics.

As a proof-of-concept experiment based on the abovementioned
factors, PZL was realized by introducing Pr3+ ions into a piezoelectric
LiTaO3 matrix as anticipated. This study revealed intense and re-
coverable PZL, which was observed by naked eye in darkness, in non-
stoichiometric LiTaO3:Pr3+ and thus broadened the family of possible
PZL materials.

2. Experimental

The LiTaO3:Pr3+ phophors were synthesized by a traditional solid-
state sintering method as follows. High purified raw materials Li2CO3
(> 99.99%), Ta2O5 (> 99.99%) and Pr2O3 (> 99.9%) were weighed in
keeping with the designed formula of Li1.00TaO3:0.01Pr3+, where the Li
concentration x was 0.97, 0.99, 1.00, 1.01, 1.03, denoted as S1 to S5 in the
following sections, and the doping concentration of Pr3+ was fixed at
1 mol%. The evenly mixed starting powders were sintered at 1070 °C
for 8 h in a common air atmosphere. The prepared phosphors were
mixed uniformly with an optical transparent epoxy resin at a total
proportion of 1:10 to form cylindrical samples (25 mm in diameter and
15 mm in thickness) for measuring PZL and afterglow (AG) perfor-
mance [26].

Phase and crystallinity of the obtained phosphors were character-
ized with an X-ray diffractometer (XRD, D/Max-2550, Rigaku, Japan)
with Cu Kα radiation (λ = 1.5418 Å) at 40 kV and 100 mA. Rietveld
analysis was performed through the powder diffraction data. Spectra of
photoluminescence (PL) were recorded through a fluorescence spec-
trophotometer (F-7000, Hitachi, Japan). Time-resolved emission spec-
trum was measured by a fluorescence spectrometer (FLS980, Edinburgh
Instruments) with a Xenon Lamb (150 W). Data of thermoluminescence
(TL) were collected via a TL meter (FU427A1, Beijing Nuclear
Instrument Factory) from room temperature to 400 °C at different
heating rates. The micro-morphology of the phosphor was character-
ized using a field emission scanning electron microscopy (FEI Nova
Nano SEM 450, America). Spectrum standing for PZL was measured by
a photon multi-channel analyzer system (QE65000, Ocean Optics, USA)
under compression. PZL curves, compression curves and AG decay
curves were recorded through a computer-controlled system that cou-
piled a photomultiplier tube (CR131, Zolix Instruments Co., China), a
photon counter (DCS103, Zolix Instruments Co., China) and a universal
testing machine (WDW-20, Shanghai Jadaronson M&C System Co.,
China). The photographs of PL, AG and PZL were captured through a
Nikon D700 camera. Before the measurement of AG, PZL and TL,
samples were optically charged via a handheld ultraviolet (UV) lamp
for 1 min and went through the delay in dark for certain time. All
measurements except TL were carried out at room temperature.

3. Results and discussion

Characterization of the phase structure and microstructure of
different LiTaO3 powder samples were accomplished by comparing the XRD spectra with the LiTaO3 standard pattern (PDF#29–0836, Fig. 1a).
No obvious peak shifts nor identified impure phases were detected in these samples. These results revealed that only single phase LiTaO3, possessing a rhombohedral symmetry and a R3c space group, existed in the phos-
phors synthesized under such preparation condition and no severe
lattice distortion was causing owing to a tiny amount of Pr3+ doping and
the slight modulation of Li concentration from 0.97 to 1.03. From an
SEM image of Li1.00TaO3:Pr3+ (sample S3, Fig. 1b), the as-synthesized
sample with high crystallization was observed to consist of crystal
grains and their sizes were nearly in uniform. The Rietveld-refined XRD
pattern of Li1.00TaO3 host lattice, was collected with a step-scanning
rate of 2° per step and a step size of 0.02 degree (Fig. 1c). The refined
lattice parameters are compared with calculated lattice parameters with
the standard lattice parameters, including a = 5.153 Å, b = 5.153 Å
and c = 13.755 Å, which suggested a greater distortion along the c axis
that arose from Pr3+ doping and/or modulation of the Li concentration
than that along the a or b axes (Table 1). With increasing Li con-
centration, the a value, which was equal to b in this typical crystal
structure, experienced a minimum at x = 1.00 and the ratio of unit cell
constants c/a rose progressively. The refined unit cell volume, which is
bigger than that of standard LiTaO3 (316.31 Å3, PDF#29–0836) in
any sample, first decreased, reached a minimum at x = 1.00, and then
increased slightly as Li concentration increased. In the previous report,
the coordination numbers of Li+ ion and Nb5+ ion are both six in
LiNbO3 crystals [45]. According to the similar crystal structure between
LiNbO3 and LiTaO3, the coordination numbers of Li+ ion and Ta5+ ion
should also be six in LiTaO3. Considering that the Pr3+ ion radius
(0.99 Å, CN = 6) is more similar to that of Li+ ion (0.76 Å, CN = 6)
and radius of Li+ ion is smaller than that of Pr3+ ion. Accordingly, the unit
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superfluous Li$^+$ ions might have entered interstitial lattice sites or Ta$^{5+}$ sites, which was speculated to be related to the damping of optical properties and unit cell volume expansion. The ideal crystal structure of LiTaO$_3$ was based on the standard pattern, which represented a typical distorted perovskite structure that was similar with that of LiNbO$_3$ in the previous report (Fig. 1d) [48,49].

Cylindrical samples were fabricated with the composition of synthesized phosphors and optical epoxy resin at certain proportions to investigate the PZL response of ferroelectric Li$_x$TaO$_3$:Pr$^{3+}$ ceramics to mechanical compression at a loading rate of 5 mm/min. Before measurement, cylindrical samples were optically charged using 254 nm radiation for 1 min and subsequent delay in darkness for 2 min to obtain PZL curves based on a relatively flat afterglow (AG) background. Typical PZL profile under compressive loading varying over time demonstrated that the intensities of stress-triggered light emission grew linearly with increased compression after the threshold (Fig. 2a and b). Interestingly, sample S1 (Li$_{0.97}$TaO$_3$:Pr$^{3+}$) was noticed to exhibit a comparatively high AG and low PZL intensities, which was not beneficial for stress sensing, whereas sample S3 (Li$_{1.00}$TaO$_3$:Pr$^{3+}$) in striking contrast showed intense PZL and low AG.

The AG decay behaviors of samples S1 and S3 were subsequently compared, which showed that the Li concentration clearly affected trap properties inside the material, which produced macroscopic variation in the intensity and duration of AG decay performance (Fig. 2a). As PZL and AG were similarly considered to originate from the evacuation process of charge carriers [50], a probable explanation for this dissimilarity involved optimized trap structure achieved by tuning the Li concentration, and the consequent high-comparison PZL was more suitable for stress imaging [51]. For all tested samples with different Li concentrations, the PZL intensity recorded at 1000 N of compressive load, with the AG intensity initially deducted, showed a positive relationship with increased of Li concentration and then underwent a concentration quenching effect (Fig. 2c). Apparently, intense PZL was obtained in samples with slightly in-excess Li concentration, at $x = 1.00$ or 1.01. Considering the refined XRD results above, increasing Li concentration caused the ratio of unit cell constants $c/a$ to progressively rise. Generally, a higher $c/a$ ratio meant better piezoelectric properties [52], which was considered here to be responsible for the present PZL enhancement. While superfluous Li$^+$ ions might enter interstitial lattice sites or Ta$^{5+}$ sites, thus affected the local crystal field around Pr$^{3+}$ ions and weakened the optical properties of the material. As a coupling-action result, a slightly Li-rich composition favored optimized PZL properties. The fusiform distribution of PZL from Li$_{1.00}$TaO$_3$:Pr$^{3+}$ was clearly observable with the naked eye, whereas it could barely be detected from Li$_{0.97}$TaO$_3$:Pr$^{3+}$ (Fig. 2c, insets i and ii). More attractively, LiTaO$_3$:Pr$^{3+}$ exhibited relatively higher PZL intensity than its isomorphic material, LiNbO$_3$:Pr$^{3+}$, under compression (Fig. 3) and the latter is a potential multipiezoelectric material in electro-mechano-optoelectronic conversion applications [27]. In recent work in our group, the PZL property of LiNbO$_3$:Pr$^{3+}$ was largely enhanced by co-doping with Gd$^{3+}$ ions [53], and thus further work is expected to find appropriate

<table>
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<th>Samples</th>
<th>Li$_{1.00}$TaO$_3$:0.01Pr$^{3+}$</th>
<th>Li$_{1.03}$TaO$_3$:0.01Pr$^{3+}$</th>
<th>Li$_{1.00}$TaO$_3$:0.01Pr$^{3+}$</th>
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<td>5.156371</td>
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<td>$b$ (Å)</td>
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<td>2.669819</td>
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<td>$R_w$ (%)</td>
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<td>$χ^2$</td>
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<td>4.36</td>
<td>3.50</td>
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Fig. 1. Phase structural and microscopic characterization of Li$_x$TaO$_3$:0.01Pr$^{3+}$ ceramics for different $x$ values: (a) XRD patterns of Li$_{1.00}$TaO$_3$:0.01Pr$^{3+}$ for $x = 0.97$, 1.00 and 1.03. (b) SEM image of Li$_{1.00}$TaO$_3$:0.01Pr$^{3+}$. (c) Rietveld refinement based on XRD data of Li$_{1.00}$TaO$_3$:0.01Pr$^{3+}$. (d) LiTaO$_3$ crystal structure.
Fig. 2. PZL performance for Li$_{1-x}$TaO$_3$:Pr$^{3+}$ with different $x$ values: (a) Luminescent curve for compressive load application to Li$_{1.00}$TaO$_3$:Pr$^{3+}$ (sample S3) over time. Inset shows the AG decay curves of specific samples recorded after 1 min excitation and 5 s delay. (b) Luminescent curve for compressive load application to Li$_{0.97}$TaO$_3$:Pr$^{3+}$ (sample S1) over time. (c) PZL intensity dependence on $x$ values, with data chosen for 1000N compressive load. Inset shows PZL photographs of Li$_{0.97}$TaO$_3$:Pr$^{3+}$ (i) and LiTaO$_3$:Pr$^{3+}$ (ii) in darkness. (d) PZL decay curve during 1st cycle of a repetitive load. (e) PZL decay curve during the 2nd cycle of a repetitive load. (f) PZL recovery performance.

Fig. 3. Comparison of PZL between LiTaO$_3$:Pr$^{3+}$ and LiNbO$_3$:Pr$^{3+}$. (a) PZL behavior of Li$_{1.00}$TaO$_3$:0.01Pr$^{3+}$. (b) PZL behavior of Li$_{1.00}$NbO$_3$:0.01Pr$^{3+}$. 
co-dopants for the LiTaO$_3$:Pr$^{3+}$ material system to produce preferable performance in real-world applications. The PZL periodic and recovery performance for a typical Li$_{1.00}$TaO$_3$:Pr$^{3+}$ sample showed that PZL intensity exhibited a notable attenuation under repeated compression-release cycles, as the corollary of gradually emptied charge carriers in traps during de-trapping processes induced by stress (Fig. 2d–f). The PZL property adequately recovered via recharging from irradiation by a UV light source. These results indicated that mildly Li-rich LiTaO$_3$:Pr$^{3+}$ phosphors emitted high-efficiency intense and recoverable PZL, which were ideal candidates for smart multifunctional probes.

The origin of the luminescent phenomenon in Pr$^{3+}$ doped LiTaO$_3$ phosphors was verified by measuring PL, AG and PZL spectra at room temperature (Fig. 4). The results showed that the prepared phosphors exhibited characteristic emissions of Pr$^{3+}$ ions with a broad spectral range under 240 nm excitation. Apparently, each sample spectrum was distinguished by different Li concentrations, revealed by the predominantly cyan- and red-emission peaks located at 511 and 615 nm respectively, which resulted in an orange-like expression of PL from samples with UV light (254 nm) excitation (Fig. 4c–i, inset of homologous image). The Commission International de L’Eclairage (CIE) chromaticity coordinates of Li$_{1.00}$TaO$_3$:Pr$^{3+}$ were calculated to be (0.349, 0.4079) using the data from the PL spectrum. These two dominant emission bands with maxima at 511 and 615 nm were ascribed to energy transitions from excited states ($^3P_0$ and $^1D_2$, respectively) to ground state ($^3H_4$) of Pr$^{3+}$ [40]. Interestingly, the optimized concentration for these two emission peaks appeared not to be coincident with the same Li concentration, and the possible explanation is speculated for varying degrees of the negative effects of defects or so-called traps-influenced recombination emission in Pr$^{3+}$ [54]. Herein, the photoluminescence excitation (PLE) spectra of material at $x = 1.00$ was used as a representative material as no obvious peak shifts were detected here at different $x$ values. Both curves, monitored at 511 and 615 nm, illustrated that the PLE spectra consisted of two prominent broad bands centered around 240 and 290 nm (Fig. 4b). The obtained PLE spectra of LiTaO$_3$:Pr$^{3+}$ were similar with those of LiNbO$_3$:Pr$^{3+}$ with high Li/Nb ratios, which could possibly be attributed to an analogous charge transfer in the regular oxyanion site [27]. Furthermore, the higher excitation was needed for LiTaO$_3$:Pr$^{3+}$ luminescence concluded to arise from the shorter PLE wavelength of LiTaO$_3$:Pr$^{3+}$ (∼240 nm here) compared with LiNbO$_3$:Pr$^{3+}$ (∼280 nm in Xu’s group [27] and our group [53]), which probably arose from the higher absorption edge of LiTaO$_3$:Pr$^{3+}$ [55].

The emission spectra of PL, AG and PZL of Li$_{1.00}$TaO$_3$:Pr$^{3+}$ at room temperature were compared, with data of AG and PZL collected after 10s and 2min delay after excitation by 254 nm for 1min, separately (Fig. 4c). A clear inconsistency was observed through this comparison under these measurement conditions, with the red-emission bands showing dominant positions in AG and PZL spectra, while cyan-
emission exhibited a significant damping compared with the PL spectrum. In consequence, the spectra of AG and PZL were in good agreement in their color expression (Figs. 2c and 4e–ii, insets). In addition, no evident peak shifting was detected, indicating the similar origin of PL, AG and PZL in LiTaO₃:Pr³⁺ from energy transitions between excited states (emission states) and ground states of Pr³⁺.

Time-resolved spectra of Li₁₀₀TaO₃:Pr³⁺ were also measured to reflect the changing course of luminescent performance in the typical material, given (Fig. 5a). All emission intensities were observed to exhibit clear attenuation as the excitation with 240 nm ceased. Comparing two dominant emission intensities that changed with time, the cyan-emission’s decay time was found to be much faster than the red-emission’s (Fig. 5b), and the spectrum was similar to the AG/PZL spectra after transitory delay (∼0.4 μs), implying that the time-resolved luminescent behavior of Li₁₀₀TaO₃:Pr³⁺ was more closely associated with its trap properties than other phosphors without AG property. A possible explanation was deduced in the following context.

More importantly, a probable mechanism was proposed to describe the luminescent processes in LiTaO₃:Pr³⁺ phosphors, as indicated by the above experimental investigations (Fig. 6a). Upon UV-light irradiation, the LiTaO₃ host absorbed energy, urging electrons excited from the valence band to the conduction band of the host. The pumped electrons subsequently transferred energy to Pr³⁺ activators through a transition level. The so-called transition level could be in the form of a charge transfer state (CTS) [27,56,57]. Pumped electrons of Pr³⁺ ions subsequently relaxed non-radiatively to emission states (3P₀, 3P₁, 3P₂, and 1D₂) and then some of them participated in PL through direct transitions from emission to ground states (3H₄ and 3H₅). Apart from electrons denoted to PL, the remaining pumped electrons were captured and trapped quasi-stably in defects that functioned as trap levels. Upon external stimulation (i.e., thermal or mechanical), the involved defects were de-trapped, supposedly favored by local piezoelectric fields caused by lattice deformation. These electrons were then released to return to emission states through direct tunneling effects [22] and de-populated to ground states as the PL process, followed by AG or PZL. Herein, the TL performance of specific samples was measured under the same excitation and delay conditions as PZL measurements to confirm the initial conjecture. TL analysis is an effective method for investigating the properties of trap levels [54,58,59], which play a significant role as transfer stations for charge carriers in persistent luminescent and ML phosphors. Generally, two key parameters of trap properties, with regard to AG/PZL, were revealed through TL curves, i.e., the trap density was proportional to the integral intensity under the TL curve and the trap depth was proportional to the corresponding TL curve peak location [50,54,59]. As anticipated, the prepared phosphors exhibited evident TL in addition to PL, AG and PZL, verifying the existence of carrier traps in the host lattice. The TL curve profile of Li₁₀₀TaO₃:Pr³⁺ was measured at a heating rate of 2 °C/s (Fig. 6b), which was further segmented into four continuous and overlapping Gaussian peaks, indicating that there were approximately four trap levels, in the coupling work, created in the synthesized material. According to Chen’s method [60], the depth of these four TL Gaussian peaks were estimated to be 0.63, 0.68, 0.96 and 1.17 eV, respectively. Comparison of the TL performance between samples S1 and S3 clearly showed that the sample S3 exhibited higher TL emission with increasing temperature than sample S1, implying that sample S3 showed superiority in the density of traps compared with sample S1 (Fig. 6c). Considering the theory mentioned above, the enhanced trap density was in favor of the de-trapping process with increasing compression. The result revealed that Li concentration strongly affected the trap density, which then influenced the luminescent properties induced by thermal or mechanical stimuli. Taking the discrepancy between PL and AG/PZL spectra into account, we stochasticly attribute it to the effect of trap distribution on the direct tunneling probabilities between trap states and different emission states in the process of AG and PZL induced by external stimulation, where the trap level energies were more closely matched with that of 1D₂ emission states compared with those of other higher-lying states [22]. Further studies should be carried out to adequately understand the underlying causation of these phenomena.

4. Conclusions

The present study revealed a novel PZL material produced through doping Pr³⁺ ions, serving as luminescent centers, into ferroelectric matrix LiTaO₃. Upon compressive loading, irradiated samples exhibited intense and recoverable light emission that could be detected by the naked eye. The observed spectral results suggested a multi-level participant process in PL, AG and PZL. XRD results demonstrated that the samples retained a typical crystal structure with rhombohedral symmetry and R₃c space group. The measurement and analysis of TL confirmed the existence of defects in the host lattice, corresponding to trap levels that were considered here as indispensable for PZL. With increased Li concentration, the ratio of the refined unit cell constants c/a increased progressively, exhibiting better piezoelectric properties. Generally, better piezoelectric properties are in favor of generating intense PZL, while superfluous Li⁺ ions would also weaken optical
properties. Owing to coupling processes and favorable trap density, slightly Li-rich nonstoichiometric phosphors performed optimally in terms of generating PZL. Finally, a mechanistic model was proposed to illustrate the probable processes of PL, AG and PZL, and to explain the similarities and differences among them. Considering the pyroelectric, piezoelectric and optoelectronic properties of LiTaO₃ host, the present examination of this PZL material broadened the prospects of LiTaO₃ materials and was expected to present great potential or extraordinary capabilities in real-time stress detection and for achieving multi-energy conversion coupled in smart devices.

Conflicts of interest

There are no conflicts of interest to declare.

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