Tunneling process in thermally stimulated luminescence of mixed $Lu_xY_{1-x}AlO_3$:Ce crystals

A. Vedda,* M. Martini, and F. Meinardi
Istituto Nazionale per la Fisica della Materia and Dipartimento di Scienza dei Materiali dell'Universita' di Milano-Bicocca, Via Cozzi 53, I-20125 Milano, Italy

J. Chval, M. Dusek, J. A. Mares, E. Mihokova, and M. Nikl
Institute of Physics, Academy of Sciences of the Czech Republic, Cukrovarnicka 10, 16253 Prague 6, Czech Republic
(Received 29 September 1999)

A thermally stimulated luminescence (TSL) study in the temperature range 20–300 °C has been performed after x-ray irradiation on mixed $Lu_xY_{1-x}AlO_3$:Ce crystals ($x=0.1-0.3$) and on $YAlO_3$:Ce. Several glow peaks have been detected, namely at 50 °C, 100–130 °C (composite structure), 175 °C and 225 °C, whose intensities are dependent upon the crystal composition. The spectrum of the emitted light features one band at 3.45 eV due to the $5d\rightarrow 4f$ transition of the Ce^{3+} ion, in good agreement with the UV excited emission spectrum. A detailed analysis of the trap depths of the peaks has been performed in the case of $Lu_{0.2}Y_{0.8}AlO_3$:Ce by partial cleaning of the glow curve. A constant energy value of approximately 1.15 eV has been found for all TSL peaks. From this result, and from the lack of a thermally stimulated current signal in the whole temperature range, it is possible to propose that TSL recombination is governed by a thermally assisted tunneling process from one trap level found at different distances from cerium emitting centers. A strong increase of TSL intensity has been observed after a prolonged annealing treatment in a vacuum atmosphere, suggesting that the electron traps are probably related to oxygen vacancies. Under this hypothesis, the relation between the frequency factors of the TSL peaks and the shortest O-Ce distances in the lattice has been investigated: an exponential dependence has been found, in accordance with what is expected for the thermally assisted tunneling recombination process.

I. INTRODUCTION

At the present time, many efforts are being dedicated to the development of new scintillators characterized by high density, fast scintillation response, and high light yield.[1] $LuAlO_3$:Ce (LuAP),[2–7] Lu_2SiO_5:Ce (LSO),[8,9] Gd_2SiO_5:Ce (GSO) (Refs. 10 and 11) all appear to be promising scintillating materials. However up to now, only the production of $YAlO_3$:Ce (YAP) has been optimized: large and homogeneous crystals (up to 15-cm long and having a diameter of ~3 cm) with Ce concentrations reaching ~0.6 at. % are grown using the Czochralski method.[12–15] Consequently, this crystal has already found application in γ-camera imaging systems: for example, a YAP:Ce multicrystal matrix detector coupled with a position sensitive photomultiplier (PSPMT) is used in γ camera for scintimammography.[16–18]

The light yield of a scintillating material should be as high as possible, and at the same time a fast response is necessary especially for medical imaging applications. Point defects both of intrinsic- or impurity-related origin can give rise to localized levels in the forbidden gap which can capture charge carriers during ionizing irradiation. The thermal depth of these levels can be monitored, for example, by thermally stimulated luminescence (TSL). The presence of such trap states can affect both the shape of the scintillation decay and the light yield of a material. Shallow traps, which give rise to TSL peaks observable below room temperature, might have decay times at room temperature (RT) comparable to or longer than the scintillation decay time, and are the cause of the slow tails observed in the scintillation decay.[19,20] On the other hand, glow peaks observed above RT are related to traps characterized by much longer decay times than the prompt scintillation. Trapping of electrons at such ''deep'' states during ionizing irradiation represents a competitive process to prompt scintillation and can lower the light yield of the material.

The TSL properties of different pure and cerium-doped orthoaluminate crystals were recently investigated.[20–24] In the case of YAP:Ce, for example, several glow peaks were found both below and above room temperature, characterized by thermal depths ranging from 0.1 to 1.4 eV.[20,22,23] A composite glow curve was detected in the TSL above RT of LuAP:Ce as well,[21–24] and thermal depths of glow peaks were found to be in the range 0.6–1.7 eV. Correlation between the presence of shallow traps below RT and the shape of the scintillation time decay was obtained in YAP:Ce;[20] comparisons between scintillation light yield and TSL above room temperature have also been performed.[20,22–24]

Very recently, mixed $Lu_xY_{1-x}AlO_3$:Ce (Lu_xY_{1-x}AP:Ce) crystals have been grown successfully, with the aim to obtain efficient scintillating crystals characterized by a higher density and light yield than YAP:Ce.[25,26] Similarly to YAP:Ce, Lu_xY_{1-x}AP:Ce mixed crystals have an orthorhombic structure and their lattice parameters are almost the same.[25] Their photoluminescence and scintillation properties are now under study;[25–27] moreover, an investigation of the trapping processes in this class of scintillating materials appears to be useful in order to understand the influence of trap levels on the scintillation mechanisms.

The aim of this paper is to study the TSL properties above RT of these newly developed mixed Lu_xY_{1-x}AP:Ce orthoaluminate crystals and to compare them with those of

TABLE I. Compositions and Ce concentrations of mixed $Lu_xY_{1-x}AP$:Ce crystals (samples for analysis were taken from the top of the pulled crystals). Stoichiometric compositions are given in italics in parentheses.

Crystal	Ce (at. %)	Lu (at. %)	Y (at. %)	Al (at. %)	O (at. %)
YAP:Ce	0.09		19.16	20.22	60.58
			(20)	*(20)*	*(60)*
$Lu_{0.1}Y_{0.9}AP$:Ce[a]	0.15	2.014	17.86	19.99	59.98
		(2)	*(18)*	*(20)*	*(60)*
$Lu_{0.2}Y_{0.8}AP$:Ce[b]	0.124	2.96	14.08	18.13	64.62
		(4)	*(16)*	*(20)*	*(60)*
$Lu_{0.3}Y_{0.7}AP$:Ce[c]	0.19	5.51	14.24	20.07	59.98
		(6)	*(14)*	*(20)*	*(60)*

[a]Almost stoichiometric composition.

[b]Excess of oxygen, lack of metal ions (Lu^{3+}, Y^{3+}, and Al^{3+}) compared to stoichiometric composition.

[c]Almost stoichiometric composition (only small excess of Y^{3+}, small lack of Lu^{3+}).

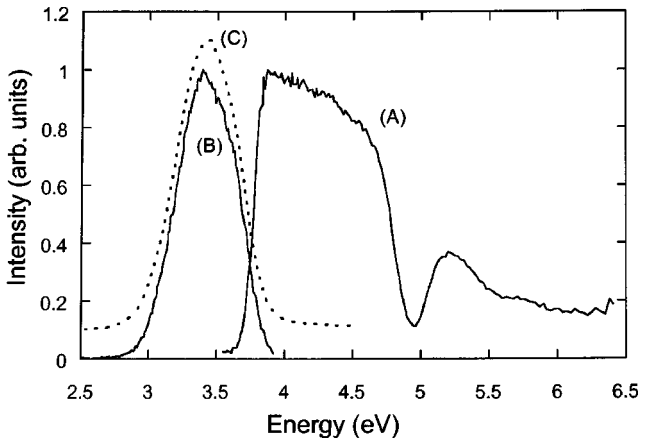

FIG. 1. UV-excited photoluminescence and TSL emission spectra of $Lu_{0.2}Y_{0.8}AP$:Ce crystal. Curve *A*, excitation spectrum at RT ($E_{em}=3.45$ eV); curve *B*, emission spectrum at RT ($E_{exc}=4.1$ eV); curve *C*, TSL spectrum integrated in the 20–200 °C temperature range.

YAP:Ce. The spectral composition of the TSL emission has been investigated; moreover, by a detailed analysis of the glow curves the evaluation of the trap depths has been obtained. Based on these data, a picture of the trap to center (Ce) recombination mechanism is presented which takes into account the existence of a thermally assisted tunneling process.

II. EXPERIMENTAL CONDITIONS

$Lu_xY_{1-x}AP$:Ce mixed orthoaluminate crystals were grown with different stoichiometry ratios ($x=0.1-0.3$) by Crytur (Turnov, Czech Republic) using the Czochralski method. A list of the crystals studied together with their compositions and Ce concentrations is presented in Table I. The Ce concentrations and compositions were evaluated by electron-beam excited x-ray analysis using a JEOL Superprobe JXA 733 electron microscope. The crystals were almost homogeneous (no cracks appeared), with dimensions of 6 cm in length and ~1.8 cm in diameter. Two of the studied $Lu_xY_{1-x}AP$:Ce samples exhibit almost stoichiometric composition while in the case of $Lu_{0.2}Y_{0.8}AP$:Ce an excess of oxygen is observed (see Table I). Samples of $7\times7\times1$ mm and $7\times7\times2$ mm were cut for TSL and photoluminescence measurements, respectively.

Photoluminescence spectra (emission and excitation) and time decays were measured in the near UV and visible range by using an Edinburgh Instruments spectrofluorometer (Model 199S), under steady-state excitation (continuous hydrogen lamp) and ns-pulsed hydrogen flash-lamp, respectively.[3] Thermally stimulated luminescence measurements after x-ray irradiation (by a Machlett OEG 50 x-ray tube operated at 32 kV) were performed from room temperature up to 300 °C with a linear heating rate of 1 °C/s using two different apparatus. In the first one the total emitted light was detected as a function of temperature by photon counting using an EMI 9635 QB photomultiplier tube. By this apparatus, simultaneous thermally stimulated current (TSC) measurements were also performed. For TSC measurements, electrodes were obtained by applying a conductive paste (JMI 4613) on the surfaces. The upper electrode was approximately 3 mm in diameter, in order to keep sufficient area for the simultaneous detection of the TSL. The TSC signal was recorded by a Keithley 617 electrometer, with an electric field of 100 V/cm; the sensitivity of the TSC measurements was of the order of 10^{-13} A. The second apparatus used was a homemade high sensitivity TSL spectrometer measuring the TSL intensity as a function of both temperature and emission wavelength. The detector was a double-stage microchannel plate followed by a 512 diode array; the dispersive element was a 140 lines/mm holographic grating, the detection range being 200–800 nm.[28] High-temperature annealing treatments were carried out in a furnace in a vacuum atmosphere (10^{-4} Torr).

III. EXPERIMENTAL RESULTS

Excitation and emission spectra of $Lu_xY_{1-x}AP$:Ce samples are characterized by wide bands in the near-UV range.[26,27] An example of these spectra is presented in Fig. 1 for $Lu_{0.2}Y_{0.8}AP$:Ce (curves *A* and *B*). The observed spectra are due to $4f\rightarrow5d$ and $5d\rightarrow4f$ allowed transitions (excitation and emission spectra, respectively); no large differences were observed for all studied mixed crystals.[27] The Ce^{3+} fast fluorescence decay constants are in the range 15–20 ns.

The TSL glow curves of the considered samples are presented in Figs. 2(a) and 2(b). Several overlapping structures are observed, whose intensities are strongly dependent upon the crystal composition: one glow peak at 50 °C is followed by a composite structure in the 100–130 °C region and by a further peak at 175 °C. A higher temperature peak at around 225 °C is observed only in the case of YAP:Ce. The spectral composition of the TSL was also investigated on all samples, showing only one emission band peaking at 360 nm (3.45 eV) which can be ascribed to the $5d\rightarrow4f$ transition of the Ce^{3+} ion. An example of a measurement is shown in Fig. 3 while the emission spectrum obtained by integrating the data

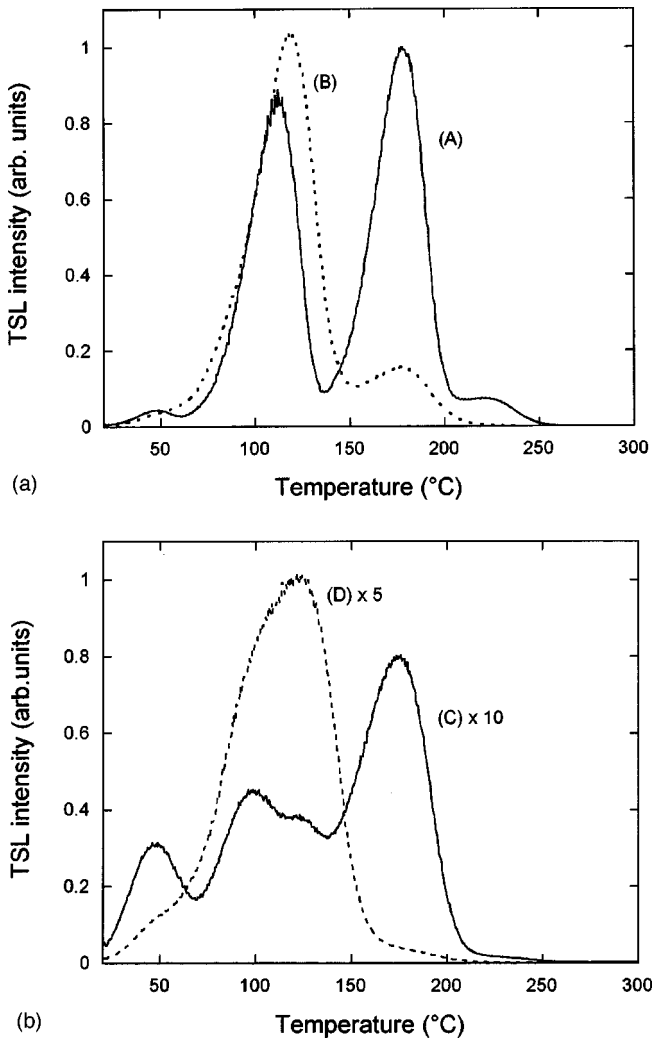

FIG. 2. (a) TSL glow curves of YAP:Ce (curve *A*) and $Lu_{0.1}Y_{0.9}AP$:Ce (curve *B*) after x-ray irradiation at RT (x-ray dose in air = 1 Gy). (b) TSL glow curves of $Lu_{0.2}Y_{0.8}AP$:Ce (curve *C*) and $Lu_{0.3}Y_{0.7}AP$:Ce (curve *D*) after x-ray irradiation at RT (x-ray dose in air = 1 Gy).

over the whole temperature range is reported in Fig. 1, curve *C*: the correspondence between the TSL and UV excited Ce^{3+} emission spectrum is very good.

In addition to the spectral composition of the emitted light, the evaluation of the thermal depths of the observed TSL peaks is worth investigation. In order to separate the individual glow peaks, TSL measurements were performed after partial heating to selected temperatures (''partial cleaning''); the data were analyzed by the initial rise method.[29] In the following, this investigation is presented in detail in the case of the $Lu_{0.2}Y_{0.8}AP$:Ce sample, in which the intensities of four main peaks (at 50 °C, 100 °C, 125 °C, and 175 °C) were comparable and so allowed a clear analysis. Figure 4 displays the Arrhenius plots of the data for a few selected temperatures while Fig. 5 shows the values of trap depths calculated by the initial rise method for all the partial cleaning temperatures considered. As can be observed, a nearly constant value is obtained from 40 °C to 165 °C, so that the same thermal depth characterizes all the four main glow peaks observed: the mean value of the data turns out to be

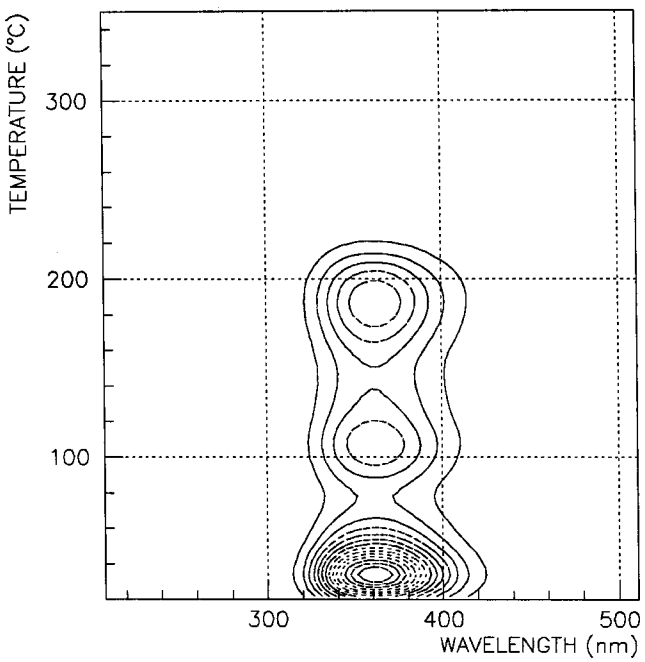

FIG. 3. Wavelength-resolved TSL measurement performed on $Lu_{0.2}Y_{0.8}AP$:Ce following x-ray irradiation at RT.

$E = 1.15 \pm 0.05$ eV. The slightly lower values of trap depth obtained below 50 °C could be due to the presence of lower temperature peaks not evident in the glow curve, which affect the shape of the initial portion of the 50 °C peak (it is worth remarking that the initial rise of the curve related to a partial cleaning at 40 °C reported in Fig. 4 starts well above the background level). The same procedure was applied to $Lu_{0.3}Y_{0.7}AP$:Ce. In this case, the ratios of the intensities of the glow peaks are such that their separation by thermal cleaning is more difficult; however, a constant energy value of about 1.2–1.3 eV was found for this crystal as well.

In order to get information about the order of kinetics of the recombination process, TSL measurements on $Lu_{0.2}Y_{0.8}AP$:Ce were performed in a wide range of doses, namely from 10^{-2} Gy to 1 Gy: a linear growth of the TSL

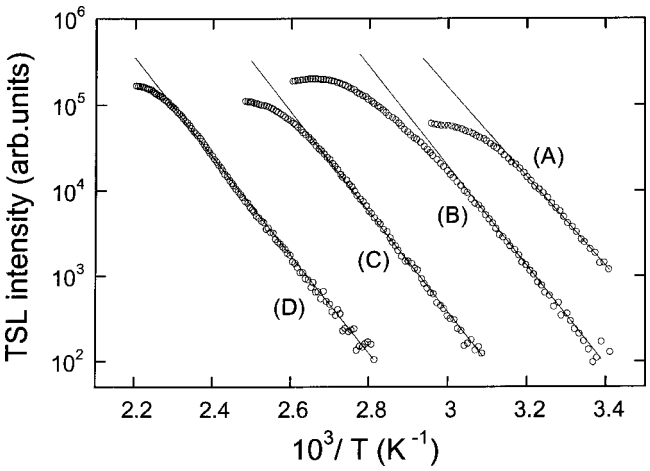

FIG. 4. Arrhenius plots of the TSL of $Lu_{0.2}Y_{0.8}AP$:Ce after partial cleaning at different temperatures: (*A*) 40 °C; (*B*) 60 °C; (*C*) 110 °C; (*D*) 165 °C.

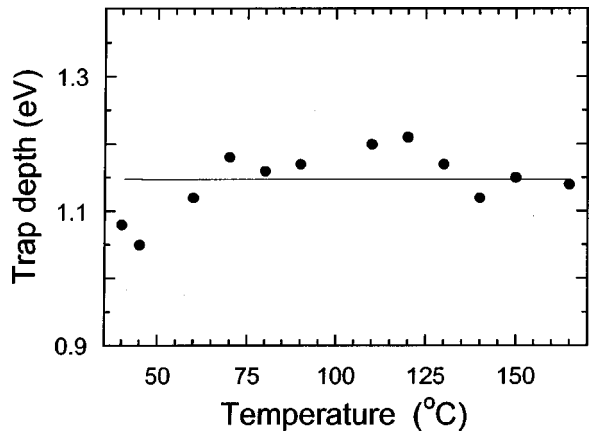

FIG. 5. Trap depth versus partial cleaning temperature in $Lu_{0.2}Y_{0.8}AP$:Ce evaluated by the initial rise method.

intensity was detected, but no shifts of the maximum temperatures of the peaks were observed, suggesting the occurrence of first-order recombination in all cases.[29] Moreover, thermally stimulated conductivity (TSC) measurements were also carried out, but no current signal was observed from RT up to 300 °C.

Finally, the last set of experiments was aimed at the investigation of the nature of point defects responsible for electron traps. A role of oxygen vacancies as electron traps can be suggested; in order to verify such possibility, the TSL of $Lu_{0.2}Y_{0.8}AP$:Ce was monitored after a prolonged (3 h) annealing in a vacuum atmosphere (10^{-4} Torr) at 1200 °C. Following this treatment, the TSL signal increases by approximately a factor 5 (Fig. 6). This result is in agreement with the above-mentioned proposal on the nature of electron traps, as the annealing in vacuum would enhance the concentration of oxygen vacancies in the crystal. However, further experimental support is needed for this hypothesis, and in this respect, electron paramagnetic resonance studies could be particularly useful.

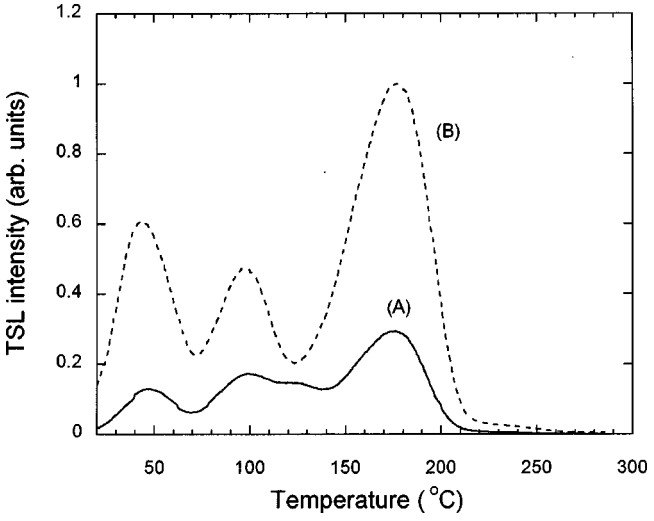

FIG. 6. Effect of a vacuum annealing treatment on the TSL glow curve of $Lu_{0.2}Y_{0.8}AP$:Ce. Curve *A*, before annealing; curve *B*, after an annealing in vacuum (10^{-4} Torr) at 1200 °C and lasting 3 h.

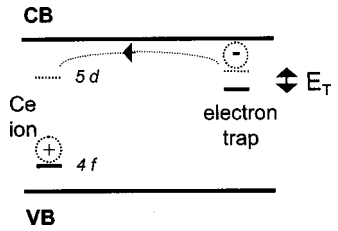

FIG. 7. Schematic representation of the TSL process in $Lu_xY_{1-x}AP$:Ce. An electron thermally freed from a trap undergoes recombination through a thermally activated tunneling mechanism with a hole localized in the $4f$ level of a Ce ion.

IV. DISCUSSION

The presented results allow us to propose a picture of the trap to center TSL recombination process occurring in $Lu_xY_{1-x}AlO_3$:Ce crystals. The spectral composition of the TSL emission leads to identify the recombination centers with cerium ions: really, capture of holes by the $4f$ level of Ce^{3+} ions with formation of Ce^{4+} is considered to occur under ionizing irradiation in several cerium activated compounds.[1] In $Lu_xAl_{1-x}O_3$:Ce, the occurrence of stable hole trapping in the $4f$ level of cerium is in agreement with the localization of this level in the forbidden gap at about 1–2 eV above the top of the valence band, as demonstrated by *x*-ray photoelectron spectroscopy studies.[30,31] Although no direct information is available in the case of $Lu_xY_{1-x}AlO_3$:Ce crystals, a similar situation could reasonably occur. Upon heating, Ce^{4+} ions can capture electrons thermally freed from traps in their $5d$ level to form an excited Ce^{3+} ion; subsequent radiative electron-hole recombination gives rise to the observed emission at 3.45 eV ($5d \rightarrow 4f$ transition) in accordance with what is observed in the UV excited emission spectra.

The presence of one single thermal energy common to all TSL peaks observed cannot be explained in the terms of the simplest TSL model, in which detrapping of electrons is followed by a transition to the conduction band, and by a subsequent recombination with holes localized at cerium luminescent sites. As a general consideration, an unambiguous interpretation of the complex pattern observed is difficult to reach. However, some hypotheses can be put forth. The results suggest the existence of direct recombination (not involving the conduction band) occurring between one trap level in different configurations (distances) with respect to the cerium centers. We propose that the mechanism enabling the recombination could be that of tunneling, in which the electron reaches the tunneling level of the trap by thermal activation (thermally assisted tunneling). Such processes have been observed in the past in other crystals such as calcite[32] and Zn_2SiO_4:Mn.[33] A schematic presentation of tunneling recombination is presented in Fig. 7. We remark that both the absence of a TSC signal and the occurrence of first-order recombination kinetics are in agreement with such a picture.

The escape probability from a trap can be written as[34]

$$P = x\nu_T \exp\left(\frac{\Delta S}{k}\right)\exp\left(\frac{-E_T}{kT}\right) = s\exp\left(\frac{-E_T}{kT}\right), \qquad (1)$$

where x is a transmission coefficient, ν_T is the thermal vibration frequency, $\exp(\Delta S/k)$ is an entropy factor, and E_T is the thermal depth of the trap. While in classical recombination processes E_T is the energy difference between the trap level and the bottom of the conduction band, in the case of thermally assisted tunneling, E_T is the energy difference between the excited (tunneling) state and the ground state of the trap. The first three terms of Eq. (1) are often unified in a single coefficient s, usually called the ''frequency factor.'' In tunneling transitions, the transmission coefficient is found to depend upon the trap to center distance in an exponential way of the form

$$x = \exp(-\phi r) \tag{2}$$

with $\phi = 2(2mI)^{1/2}/\hbar$, where m is the electron mass, $\hbar$ is the Plank's constant, and I is the energy difference between the excited tunneling trap state and the potential barrier of the recombination site.[29] In this frame, the existence of several discrete traps to center distances can lead to different glow peaks characterized by the same thermal energy E_T and different frequency factors s.

Based on the existence of first-order recombination kinetics, the frequency factor s for a peak of maximum temperature T_m can be estimated by the equation:

$$\frac{\beta E_T}{kT_m^2} = s \exp\left(\frac{-E_T}{kT_m}\right), \tag{3}$$

where β is the heating rate (and E_T is the thermal energy). This expression, derived by Hoogenstraten[35] for classical first-order recombination through the conduction band, was later found to be valid also in the case of thermally assisted tunneling,[36] assuming that in this case E_T is the difference between the excited (tunneling) state and the ground state of the trap.

The frequency factors calculated using Eq. (3) and $E_T = 1.15$ eV turn out to be 1×10^{17}, 3×10^{14}, 3×10^{13}, and $5\times10^{11}\,s^{-1}$ for the 50, 100, 125, and 175 °C peaks of $Lu_{0.2}Y_{0.8}AP$:Ce, respectively. In this calculation the maximum temperature of the fourth peak was taken at 178 °C: in fact, this was the value obtained in the glow curves after partial cleanings above 140 °C, in which this glow peak appeared by itself. We emphasize the qualitative character of this evaluation in the case of lower temperature peaks, whose maximum temperatures could be slightly affected by their superposition.

Following the hypothesis that electron traps are related to oxygen vacancies, the existence of tunneling recombination between one trap level and cerium ions at different distances was checked by studying the relation between the calculated frequency factors and the O-Ce distances in the lattice. Actually, by taking into account the ionic radius of Ce and those of the cations of the host matrix [r(Ce)=1.04 Å, r(Y)=0.9 Å, r(Lu)=0.85 Å, and r(Al)=0.51 Å] the localization of Ce ions at Y (Lu) substitutional sites is expected, rather than at Al lattice sites. Electron paramagnetic resonance (EPR) studies of pure YAP:Ce crystals confirm this hypothesis;[37] in mixed crystals, preliminary EPR results confirm that Ce ions can be localized at both Y and Lu lattice positions.[38]

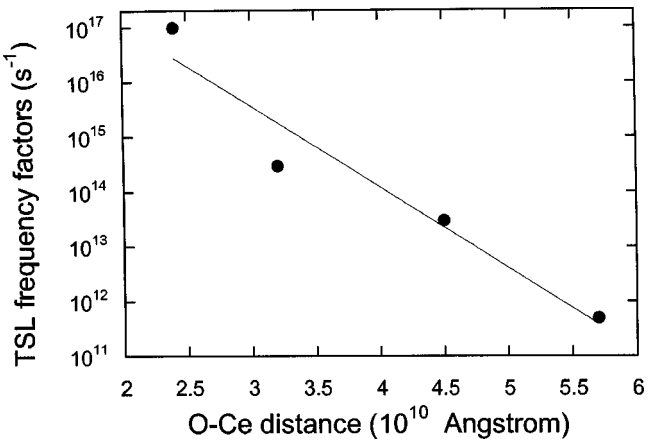

FIG. 8. Frequency factors of the TSL peaks of $Lu_{0.2}Y_{0.8}AP$:Ce vs the nearest O-Ce distances. The continuous line is the exponential fit performed on the data.

O-Y distances within a few angstroms from oxygen sites were calculated in a YAP crystal by using structural data.[39] Lattice parameters of mixed crystals have also been determined recently by x-ray diffraction[25] and comparison of these data shows that only small differences occur (for $Lu_{0.2}Y_{0.8}AP$:Ce crystals $a=5.17$ Å, $b=5.33$ Å, and $c=7.36$ Å while for YAP $a=5.18$ Å, $b=5.33$ Å, and $c=7.38$ Å). Taking into account the above-mentioned localization of Ce ions at Y sites, the calculated O-Y distances were considered representative of O-Ce distances, too. The relation between the frequency factors and the distances of the four nearest neighbors is shown in Fig. 8: in this figure, the abscissa values were obtained by averaging ion distances of Ref. 39 differing by 0.2 Å or less. In spite of the already mentioned qualitative character of the evaluation of the frequency factors, to a good approximation an exponential dependence is observed over several decades, in agreement with Eq. (2): this fact supports the proposed model of thermally assisted tunneling from one trap level to cerium sites. Actually, a deeper insight concerning this mechanism, including a comparison with the classical one (trap-center interaction via conduction band) could be obtained on samples by varying Ce concentrations starting from Ce traces up to the concentrations considered in the present investigation.

V. CONCLUSIONS

This investigation on the TSL properties of mixed $Lu_xY_{1-x}AP$:Ce crystals has allowed us to put in evidence the existence of several peaks in the glow curve in the temperature range 20–300 °C; however, their thermal depths, evaluated by the initial rise method, were found to be similar: a nearly constant value of approximately 1.15 eV was obtained for all peaks. This result was considered together with a number of other experimental data as the analysis of the spectral emission of the TSL, TSC measurements, the study of the dependence of the signals upon dose and annealing treatment. The overall picture calls for the existence of localized trap-to-center (Ce) recombination through a thermally assisted tunneling process. The nature of the defects responsible for trap levels is still matter of debate: however,

the results obtained after prolonged annealing in reducing atmosphere support the hypothesis that such defects can be related to oxygen vacancies. Under this assumption, an exponential relation between the frequency factors of the peaks and the nearest O-Ce distances in the lattice was found, as expected in the frame of the above-mentioned thermally assisted tunneling model.

ACKNOWLEDGMENTS

This work was partly supported by the IP research program of the Swiss National Science Foundation (SNSF), Grant No. 7 IP 051218. The authors are grateful to the members of the Crystal Clear Collaboration (CERN RD18 project) for fruitful discussions.

*Author to whom correspondence should be addressed. FAX: 39-02-64485400.

Electronic address: Anna.Vedda@mater.unimib.it

[1] P. A. Rodnyi, *Physical Processes in Inorganic Scintillators* (Chemical Rubber, London, 1997).

[2] M. Moszynski, D. Wolski, T. Ludziejewski, M. Kapusta, A. Lempicki, C. Brecher, D. Wisniewski, and A. J. Wojtowicz, Nucl. Instrum. Methods Phys. Res. A **385**, 123 (1997).

[3] J. A. Mares, M. Nikl, J. Chval, I. Dafinei, P. Lecoq, and J. Kvapil, Chem. Phys. Lett. **241**, 311 (1995).

[4] C. Dujardin, C. Pedrini, P. Meunier-Beillard, B. Moine, J. C. Gacon, and A. Petrosyan, J. Lumin. **72–74**, 759 (1997).

[5] C. Dujardin, C. Pedrini, W. Blanc, J. C. Gacon, J. C. Van't Spijker, O. W. V. Frijns, C. W. E. van Eijk, P. Dorenbos, R. Chen, A. Fremount, F. Tallouf, S. Tavernier, P. Bruyndonckx, and A. G. Petrosyan, IEEE Trans. Nucl. Sci. **43**, 467 (1998).

[6] D. Wisniewski, A. J. Wojtowicz, and A. Lempicki, J. Lumin. **72–74**, 789 (1997).

[7] C. Dujardin, C. Pedrini, J. C. Gacon, A. G. Petrosyan, A. N. Belsky, and A. N. Vasil'ev, J. Phys.: Condens. Matter **9**, 5229 (1997).

[8] R. Visser, C. L. Melcher, J. S. Schweitzer, H. Suzuki, and T. A. Tombrello, IEEE Trans. Nucl. Sci. **41**, 689 (1994).

[9] J. Adriessen, A. Sobolev, A. Kuznetsov, H. Merenga, P. Dorenbos, and C. W. E. van Eijk, in *Proceedings of the International Conference on Inorganic Scintillators and their Applications (SCINT95), Delft, The Netherlands, 1995*, edited by P. Dorenbos and C. W. E. Van Eijk (Delft University Press, Delft, 1996), p. 130.

[10] C. L. Melcher, J. S. Schweitzer, T. Utsu, and S. Akiyama, IEEE Trans. Nucl. Sci. **37**, 161 (1990).

[11] H. Ishibashi, K. Kurashige, Y. Kurata, K. Susa, M. Kobayashi, M. Tanaka, K. Hara, and M. Ishii, IEEE Trans. Nucl. Sci. **45**, 518 (1998).

[12] M. Kapusta, J. Pawelke, and M. Moszynski, Nucl. Instrum. Methods Phys. Res. A **404**, 413 (1998).

[13] M. Moszynski, M. Kapusta, D. Wolski, W. Klamra, and B. Cederwall, Nucl. Instrum. Methods Phys. Res. A **404**, 157 (1998).

[14] S. Baccaro, K. Blazek, F. de Notaristefani, P. Maly, J. A. Mares, R. Pani, R. Pellegrini, and A. Soluri, Nucl. Instrum. Methods Phys. Res. A **361**, 209 (1995).

[15] J. A. Mares, M. Nikl, C. Pedrini, B. Moine, and K. Blazek, Mater. Chem. Phys. **32**, 342 (1992).

[16] R. Pani, R. Pellegrini, A. Soluri, G. De Vincentis, R. Scafe, and A. Pergola, Nucl. Instrum. Methods Phys. Res. A **409**, 524 (1998).

[17] S. Majewski, F. Farzanpay, A. Goode, B. Kross, D. Steinbach, A. Weisenberger, M. Williams, and R. Wojcik, Nucl. Instrum. Methods Phys. Res. A **409**, 520 (1998).

[18] L. H. Barone, K. Blazek, D. Bollini, A. Del Guerra, F. de Notaristefani, G. De Vincentis, G. di Domenico, M. Galli, M. Giganti, P. Maly, R. Pani, R. Pellegrini, A. Pergola, A. Piffanelli, F. Scopinaro, A. Soluri, and F. Vittori, Nucl. Instrum. Methods Phys. Res. A **360**, 302 (1995).

[19] M. Nikl, K. Nitsch, P. Bohacek, M. Martini, E. Mihokova, A. Vedda, S. Croci, G. P. Pazzi, P. Fabeni, S. Baccaro, B. Borgia, I. Dafinei, M. Diemoz, G. Organtini, E. Auffray, P. Lecoq, M. Kobayashi, M. Ishii, and Y. Usuki, Appl. Phys. Lett. **71**, 3755 (1997).

[20] A. J. Wojtowicz, J. Glodo, A. Lempicki, and C. Brecher, J. Phys.: Condens. Matter **10**, 8401 (1998).

[21] D. Wisniewski, W. Drozdowski, A. J. Wojtowicz, A. Lempicki, P. Dorenbos, J. T. M. de Haas, C. W. E. Van Eijk, and A. J. J. Bos, Acta Phys. Pol. A **90**, 377 (1996).

[22] R. H. Bartram, D. S. Hamilton, L. A. Kappers, and A. Lempicki, J. Lumin. **75**, 183 (1997).

[23] A. J. Wojtowicz, J. Glodo, W. Drozdowski, and K. R. Przegietka, J. Lumin. **79**, 275 (1998).

[24] A. J. Wojtowicz, W. Drozdowski, D. Wisniewski, K. Wisniewski, K. R. Przegietka, H. L. Oczkowski, and T. M. Piters, Radiat. Meas. **29**, 323 (1998).

[25] J. Chval, D. Clement, J. Giba, J. Hybler, J.-F. Loude, J. A. Mares, E. Mihokova, C. Morel, K. Nejezchleb, M. Nikl, A. Vedda, and H. Zaidi, Nucl. Instrum. Methods Phys. Res. A (to be published).

[26] J. A. Mares, M. Nikl, J. Chval, E. Mihokova, K. Nejezchleb, J. Giba, D. Clement, J.-F. Loude, and C. Morel (unpublished).

[27] J. A. Mares, M. Nikl, J. Chval, K. Nejezchleb, J. Giba, D. Clement, J.-F. Loude, and C. Morel, Radiat. Eff. Defects Solids **150**, 59 (1999).

[28] M. Martini, S. Paravisi, and C. Liguori, Radiat. Prot. Dosim. **66**, 447 (1996).

[29] S. W. S. McKeever, *Thermoluminescence of Solids*, Cambridge Solid State Science Series (Cambridge University Press, Cambridge, U.K., 1985).

[30] C. Pedrini, D. Bouttet, and C. Dujardin, *Proceedings of the International Conference on Inorganic Scintillators and their Applications (SCINT95), Delft, 1996* (Ref. 9), p. 103.

[31] D. Bouttet, C. Dujardin, C. Pedrini, W. Brunat, D. Tran Minh Duc, and J. Y. Gesland, *Proceedings of the International Conference on Inorganic Scintillators and their Applications (SCINT95), Delft, 1996*, (Ref. 9), p. 111.

[32] R. Visocekas, T. Ceva, C. Marti, F. Lefaucheux, and M. C. Robert, Phys. Status Solidi A **35**, 315 (1976).

[33] P. Avouris and T. N. Morgan, J. Chem. Phys. **74**, 4347 (1981).

[34] D. Curie, *Luminescence in Crystals* (Wiley, New York, 1960).

[35] W. Hoogenstraaten, Philips Res. Rep. **13**, 515 (1958).

[36] I. F. Chang and P. Thioulouse, J. Appl. Phys. **53**, 5873 (1982).

[37] H. R. Asatryan, J. Rosa, J. A. Mares, Solid State Commun. **104**, 5 (1997).

[38] H. R. Asatryan (private communication).

[39] G. Bergerhoff, R. Hundt, R. Sievers, and I. D. Brown, J. Chem. Inf. Comput. Sci. **23**, 66 (1983).