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# Positron annihilation in samarium

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**Abstract.** The positron annihilation characteristics of deformed and annealed samarium have been investigated. The temperature dependence of the lineshape parameter $S$ observed for deformed samples is described by a model of competing positron trapping in dislocations and vacancy-like defects. The results indicate that the positron trapping in deformed samples is controlled by dislocations. Positron detrapping from dislocations appears not to be responsible for the positive temperature dependence of the parameter $S$. The analysis of the experimental data yields a positron–dislocation binding energy of 18 meV. The recovery of the deformation damage takes place through the temperature range 373–703 K. A value of $(199 \pm 2)$ ps is attributed to the bulk positron lifetime.

## 1. Introduction

Positron annihilation measurements in rare earth metals were performed at the beginning of the application of this technique to investigate solids [1]. However, it appears that positron annihilation spectroscopy (PAS) has been barely applied to investigate defects in these materials. In recent years, the literature has reported positron annihilation data for cerium and gadolinium only [2–5]. This lack of positron annihilation investigations in rare earth metals can be attributed to the low technological interest in these materials in the past.

At present, the magnetic characteristics of some of these metals have raised their interest. Among them, samarium and gadolinium are the most important. PAS has been applied to investigate the recovery of deformation damage in gadolinium [3], but so far similar investigations have not been reported for samarium, to our knowledge. Moreover, the characteristics of the lattice defects in samarium appear not to be well established.

On the other hand, the local density approximation (LDA) calculations of the bulk positron lifetime for cerium and gadolinium disagree noticeably with the experimental values, independently of the method used to evaluate the enhancement term describing the screening charge around the positron [6–8]. The values calculated using a total enhancement factor for core and valence electrons together show much larger discrepancies with the experimental values than those obtained calculating separate enhancement factors. However, the LDA calculations have demonstrated that it is not necessary to evaluate the enhancement factor for core and valence electrons separately, except for cerium and gadolinium [6–8]. Positron lifetime measurements should be performed on the remaining lanthanides to elucidate whether these exceptions are a particular characteristic of the 4f rare earth metals. The investigation of the positron annihilation characteristics in samarium can give insight into this point and provide useful data about the defect recovery in this metal.

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## 2. Experimental method

The samples were prepared from a samarium plate of 99.9 at.% purity supplied by the Institute of Physics of Solids and Semiconductors, Academy of Sciences of BSSR (Minsk). The plate, 5 mm thick, was uniaxially compressed at room temperature up to a thickness reduction of 50%. The samples were carefully polished until mirror-like surfaces were achieved. Doppler broadening measurements at low temperatures were performed on a pair of deformed samples set into a closed He cycle cryostat. Afterwards, this pair was isochronally annealed for 30 min up to 763 K in 30 K steps. To avoid oxidation and sublimation, these annealings were carried out in a vacuum of $10^{-3}$ Pa, i.e. the samarium vapour pressure at 781 K. Positron lifetime measurements at room temperature were made after each annealing. After annealing at 763 K, Doppler broadening measurements at low temperature were performed again. Another pair of deformed samples was annealed for 30 min at 923 K in vacuum and its positron lifetime spectrum measured at room temperature. For low-temperature measurements, the temperature was selected arbitrarily going up and down.

The Doppler broadening measurements were made with a zero- and gain-stabilized high-purity Ge detector having an energy resolution of 1.62 keV at the 1.33 MeV line of $^{60}Co$. For lifetime measurements a spectrometer with a time resolution of 225 ps (FWHM) was used. The positron source was $^{22}Na$ inside sealed Kapton foils. The Doppler broadening of the annihilation peak was characterized by the lineshape parameter $S$, defined as the fraction of counts within an energy window of 1.50 keV centred at 511 keV. The lifetime spectra were analysed with the programs RESOLUTION and POSITRONFIT.

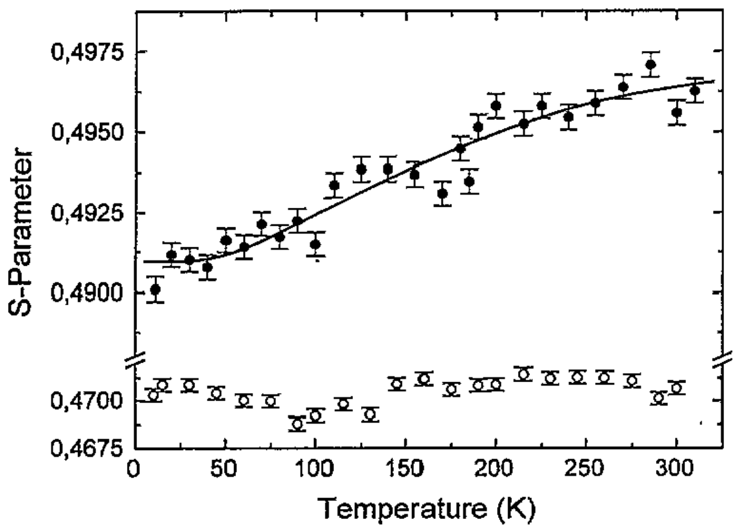


**Figure 1.** Lineshape parameter as a function of temperature for deformed (•) and annealed (o) samarium. The curve represents the least-squares fit of the points to equation (1).

## 3. Results and discussion

Figure 1 depicts the parameter $S$ as a function of temperature for the same pair of samples, after deformation and after the last isochronal annealing. The $S$ values show a reversible

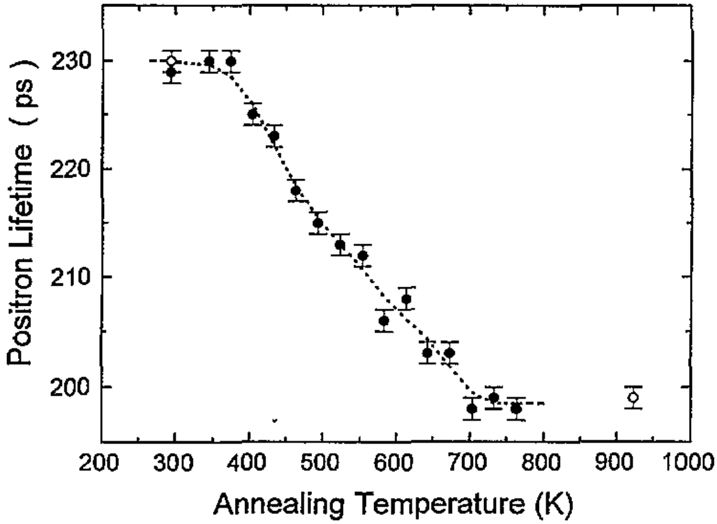


**Figure 2.** Positron lifetime versus annealing temperature for deformed samarium. Isochronally (•) and isothermally (○) annealed samples.

behaviour with temperature for both cases. As-deformed samples show a light increase of the $S$ values with temperature. However, this temperature dependence disappears after annealing at 763 K. After subtracting background and the source component, the lifetime spectra only could be analysed in terms of a single exponential component. These lifetime spectra were characterized by the single positron lifetime $\tau = \lambda^{-1}$, where $\lambda$ is the decay rate of the single exponential term. The values after isochronal annealing are shown in figure 2 along with the results for a pair of samples isothermally annealed at 923 K. A continuous recovery of the value is observed over the temperature range 373–703 K. Through this wide recovery stage, the positron lifetime spectra could not be consistently decomposed into two spectral components. This is interpreted as evidence of the presence of a complex structure of defects acting as positron traps in deformed samarium.

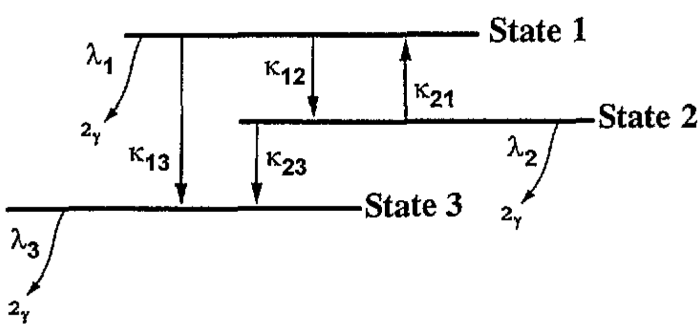


**Figure 3.** Three-state trapping model including detrapping from shallow traps (state 2) and transitions from shallow traps to deep traps (state 3). State 1 represents the bulk state.

The temperature dependence of the $S$ values, shown in figure 1, indicates that there is no trapping saturation for deformed samples annealed at $T \leqslant 373$ K, and therefore the value of $(230 \pm 2)$ ps would not correspond to the lifetime for positrons trapped in vacancy-like defects or dislocations. The characteristic parameters for positrons in these types of defect are expected to be temperature independent. The increase of the parameter $S$ with temperature cannot be attributed to volume expansion because it is not observed after annealing at 763 K. Thus, it should be attributed to a temperature-dependent competing trapping in different types of defect. The observed $\tau$ and $S$ values would represent a weighted mean value for positrons in the bulk and positrons trapped into defects produced by plastic deformation. It is expected that vacancies in samarium should be mobile below room temperature as happens in other lanthanides [9], so that, during deformation at room temperature, vacancies can be trapped by dislocations and impurities. The presence of tridimensional vacancy clusters, or voids, can be rejected because the lifetime spectra do not show a long-lived component attributable to them. The defects responsible for the trapping should be dislocations and a wide variety of vacancy-like defects associated with dislocations such as a vacancy trapped at a dislocation (i.e. a single jog), jogs, vacancy–impurity pairs trapped at dislocations, and so on. Perfect dislocations or straight parts of dislocations would act as shallow positron traps, while the vacancy-like defects would be deep traps.

To obtain information from the temperature dependence of the parameter $S$, we have applied the method recently proposed by Trumpy for describing the positron trapping in deformed nickel [10]. Figure 3 depicts the annihilation scheme. This method, based on the model of Smedskjaer *et al* for positron trapping in dislocations [11], results in a parameter $S$ given by (see appendix)

$$S = S_1 + (S_3 - S_1)\, P_3 \tag{1}$$

where $S_1$ is the value of the annihilation parameter for the material free of defects, $S_3$ the characteristic value for vacancy-like defects present in the deformed samples, and $P_3$ is the annihilation probability for positrons in deep traps given by equation (A7). $P_3$ is a function of the transition rates $\kappa_{ij}$ and of the positron annihilation rate in the bulk $\lambda_1$.

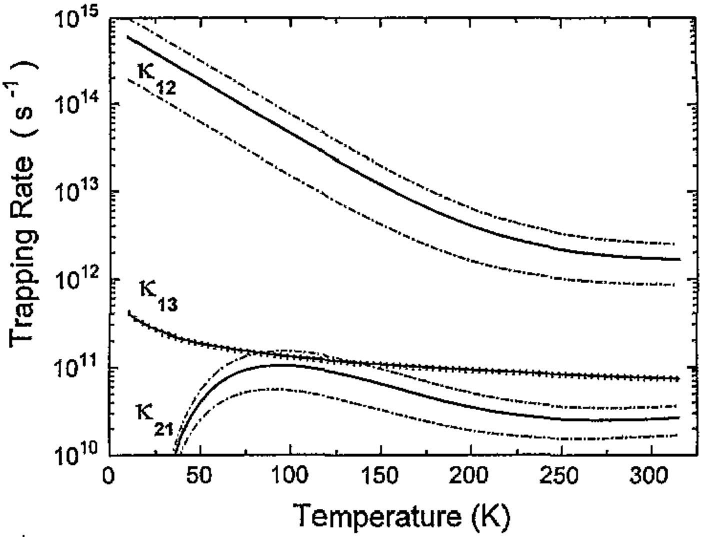


**Figure 4.** Positron trapping and detrapping rates as a function of temperature in deformed samarium. Curves $\kappa_{12}$ and $\kappa_{13}$ represent the trapping rate for dislocations and vacancy-like defects, respectively. Curve $\kappa_{21}$ corresponds to the detrapping rate from dislocations. The curves are given by equations (A8). Dashed curves represent the confidence intervals for the $\kappa_{ij}$ values.

From the best fit of the $S$ values to equation (1) we obtained the following values for the adjustable parameters: $E_b = (18 \pm 1)$ meV, $\kappa_{23} < 10^7$ s$^{-1}$, $\rho = (6.6 \pm 0.4) \times 10^{15}$ m$^{-2}$, $\gamma = (29 \pm 4) \times 10^{-3}$ K$^{-1}$, $\nu_1 = (2.4 \pm 1.0) \times 10^{-4}$ m$^2$ s$^{-1}$, $\mu = (530 \pm 100)$, $b = (1.32 \pm 0.08) \times 10^{12}$ K$^{1/2}$ s$^{-1}$, $S_1 = (0.4909 \pm 0.0010)$, and $S_3 = (0.5170 \pm 0.0016)$. It should be noted that the fit was optimized using $a \simeq 0$ s$^{-1}$. The value $\lambda_1 = 5.03 \times 10^9$ s$^{-1}$ obtained from positron lifetime measurements of well annealed samples is used for the fit. These parameters are defined in the appendix.

The above values are comparable to those obtained by Trumpy for nickel [10], except for $\nu_1$ and $\mu$ that are two and one orders of magnitude higher, respectively. The last is consistent with a low positron–dislocation binding energy of 18 meV against the value of 38 meV found by Trumpy for nickel. According to the model of Smedskjaer *et al*, the lower the binding energy, the stronger the temperature dependence of the specific trapping rate for dislocations and the higher its value [11]. Figure 4 depicts the temperature dependence of the trapping rates for dislocations $\kappa_{12}$ and vacancy-like defects $\kappa_{13}$, and the detrapping rate for dislocations $\kappa_{21}$, along with their respective confidence intervals. These curves are given by equations (A8) using the parameters obtained from the fit. The $\kappa_{12}$ is two or three orders of magnitude higher than $\kappa_{13}$ over the temperature range 10–310 K, while the detrapping rate $\kappa_{21}$ remains lower than $\kappa_{13}$ continuously.

The recovery through the range 373–703 K is attributed to successive annealing out of vacancy-like defects and dislocations. The lifetime value of $(199 \pm 2)$ ps found after annealing at $T > 703$ K is ascribed to positrons in the bulk. This value disagrees with the value of 246 ps reported by Rodda and Stewart [1], but agrees with the value of 202 ps predicted by Welch and Lynn [12].

## 4. Conclusions

It is concluded that the positron annihilation characteristics found in deformed samarium can be described by a temperature-dependent competing trapping in dislocations and vacancy-like defects. The positive temperature dependence of the parameter $S$ found for deformed samarium is consistent with positron trapping controlled by dislocations. Positron detrapping from dislocations appears to be unimportant in determining this temperature dependence.

Finally, it should be mentioned that the paramagnetic–antiferromagnetic transition taking place for samarium at 105 K does not have any meaningful effect on the positron lifetime and the parameter $S$.

## Appendix

A scheme of the trapping model used above is shown in figure 3. The rate equations of the model are

$$\begin{cases} \dot{n}_1(t) = -(\lambda_1 + \kappa_{12} + \kappa_{13})n_1(t) + \kappa_{21}n_2(t) \\ \dot{n}_2(t) = -(\lambda_2 + \kappa_{21} + \kappa_{23})n_2(t) + \kappa_{12}n_1(t) \\ \dot{n}_3(t) = -\lambda_3 n_3(t) + \kappa_{13}n_1(t) + \kappa_{23}n_2(t) \end{cases} \tag{A1}$$

where 1 is the bulk state, 2 and 3 the bound states in shallow and deep traps, respectively, $n_i$ is the fraction of positrons in the $i$ state, $\lambda_i$ the annihilation rate, and $\kappa_{ij}$ the transition rate from state $i$ to state $j$.

By imposing the initial conditions

$$n_1(0) = 1$$

$$n_2(0) = n_3(0) = 0$$

the positron fraction in the sample, $n(t)$, would be given by

$$n(t) = n_1(t) + n_2(t) + n_3(t) = I_+ e^{-\lambda_+ t} + I_- e^{-\lambda_- t} + I_3 e^{-\lambda_3 t}. \tag{A2}$$

Here $\lambda_+$ and $\lambda_-$ are defined as

$$\lambda_\pm = \frac{\alpha + \beta \pm \sqrt{(\alpha - \beta)^2 + 4\kappa_{12}\kappa_{21}}}{2} \tag{A3}$$

with $\alpha = \lambda_1 + \kappa_{12} + \kappa_{13}$ and $\beta = \lambda_2 + \kappa_{21} + \kappa_{23}$. The mean positron lifetime, $\bar{\tau} = I_+\tau_+ + I_-\tau_- + I_3\tau_3$, is also given by

$$\bar{\tau} = \sum_{i=1}^{3} P_i \tau_i \tag{A4}$$

where $P_i = \int_0^\infty \lambda_i n_i(t)\,dt$ is the probability that the positron annihilates in the $i$ state, and $\tau_i = \lambda_i^{-1}$ is the lifetime.

A similar expression to equation (A4) gives the lineshape parameter $S$:

$$S = \sum_{i=1}^{3} P_i S_i \tag{A5}$$

where $S_i$ is the lineshape parameter characteristic of positrons in the $i$ state. Assuming the approximation $S_2 \approx S_1$ proposed by Smedskjaer *et al* [11], the parameter $S$ could be expressed as

$$S = (1 - P_3)S_1 + P_3 S_3 \tag{A6}$$

where the annihilation probability for positrons in deep traps $P_3$ is given by

$$P_3 = \frac{\kappa_{12}\kappa_{23} + \kappa_{13}\ (\kappa_{21} + \kappa_{23} + \lambda_1)}{\kappa_{13}\kappa_{21} + \kappa_{12}\kappa_{23} + \kappa_{13}\kappa_{23} + (\kappa_{12} + \kappa_{13} + \kappa_{21} + \kappa_{23})\ \lambda_1 + \lambda_1^2}. \tag{A7}$$

The following arguments would justify the approximation $S_2 \approx S_1$. According to calculations performed on f.c.c. metals, the positron binding energy for pure dislocations $E_b$ is in the range 0.02–0.10 eV. This results in an angular correlation function and a lifetime for positron annihilation at dislocations that are indistinguishable experimentally from those for the bulk [13, 14]. For rhombohedral samarium, the $E_b$ value may be similar, or even lower, because its dislocation core structure is expected to be wider than the one calculated for f.c.c. metals.

The temperature dependence for the transition rates can be expressed according to [10, 11] as

$$\begin{aligned}
\kappa_{13} &= a + bT^{-1/2} \\
\kappa_{12} &= \rho\nu_1 \left[1 + \mu\ \exp(-\gamma T)\right] \\
\kappa_{21} &= \frac{m^* k_B}{4\hbar^2\pi^{1/2}} T \left(\frac{E_b}{k_B T}\right)^{-1/2} \exp\left(\frac{-E_b}{k_B T}\right) \nu_1 \left[1 + \mu\ \exp(-\gamma T)\right]
\end{aligned} \tag{A8}$$

where $a$, $b$, $\gamma$, $\nu_1$ and $\mu$ are constants, $\rho$ the dislocation density, $E_b$ the positron binding energy to the dislocation, and $m^*$ the effective mass of the positron.

The transition rate $\kappa_{23}$ is assumed temperature independent.

Now, the temperature dependence of the parameter $S$ would be obtained introducing equations (A8) into equation (A7).

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