Lifetime of Positrons Trapped by F-Centers in KCl.

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In the following letter account is given of a series of experiments on the lifetime distribution of positrons annihilating in nonstoichiometric KCl single crystals colored by F-centers in concentrations of the order of 10^{17} centers per cm^3. The role played by crystal defects in determining the fate of positrons in an alkali halide crystal was conjectured by GOL'DANSKII and PROKOP'EV [1], with explicit reference to the possibility of the formation of a Ps-like system in voids created by ionic vacancies. As regards stoichiometric crystals, only two experiments intended to study the effectiveness of crystal defects on the annihilation spectrum have been reported [2,3]. In this case, it is believed that positive ion vacancies have the highest positron capture cross-section [4]. Quite recently, HERLACH and HEINRICH [5] revealed a low-momentum component in the angular distribution of γ-rays emitted by positrons annihilating in additively colored KCl. This component was attributed to positrons trapped in F-centers.

In the present experiment, positron lifetime spectra are measured both for additively and electrolitically colored KCl. Heat treatments were made in order to obtain the highest F-center concentration, or, alternatively, a partial conversion to colloidal centers (X-centers, following the nomenclature adopted by SCHULMANN and COMPTON [6]): in the former case, heating at 550 °C for (75 ÷ 90) s followed by quenching by contact with a cold copper mass; in the second case, heating at 300 °C for 90 min followed by quenching. After the heat treatment, the crystals (two small slabs, approximately $(10 \times 10 \times 1.5)$ mm^3) were mounted with a ^{22}N positron source on mica foils (0.7 mg/cm^2

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[1] V. I. GOL'DANSKII and E. P. PROKOP'EV: *Fiz. Tver. Tela*, **6**, 3301 (1964) (English translation: *Sov. Phys. Solid State*, **6**, 2641 (1965)).

[2] W. BRANDT, H. F. WAUNG and P. LEVY: in *Proceedings of the International Symposium on Color Centers in Alkali Halides, Rome, 1968* (unpublished).

[3] M. BERTOLACCINI and A. DUPASQUIER: *Phys. Rev.* (1970) (to be published).

[4] W. BRANDT: in *Proceedings of the Wayne Conference on Positron Annihilation*, edited by A. T. STEWART and L. O. ROELLIG (New York, 1967).

[5] D. HERLACH and F. HEINRICH: *Phys. Lett.*, **31 A**, 47 (1969).

[6] J. H. SCHULMANN and W. D. COMPTON: *Color Centers in Solids* (Oxford, 1962).

thick), and wrapped in light-proof aluminum foil. The entire manipulation was conducted in subdued red light. The lifetime spectrum was then measured with the standard method described in previous papers (see ref. [3]); the prompt curve of the timing apparatus has a F.W.H.M. of $3.0 \cdot 10^{-10}$ s, measured with ^{60}Co γ-rays. Immediately after the time measurement, the optical absorption of a thin slice cut from the crystal sample was determined in a Hitachi Perkin-Elmer EPS-3T spectrophotometer. Smakula's equation [7] was applied to calculate F-center concentration. The accuracy of the obtained concentration values is estimated to be of the order of 10%, errors being mainly due to the incomplete homogeneity in coloration, and to the need to read optical densities which were sometimes very high in relation to the spectrophotometer sensitivity.

The effect of coloring on positron annihilation is a marked distortion of the time spectrum. At least three exponentially decaying components are found, whereas only two components exist in a stoichiometric crystal [3,8]. The experimental results are collected in Table I: the components in this Table are numbered in order of increasing mean life.

TABLE I. – *Mean lives and intensities of positrons annihilating in colored* KCl.

F-center concentration (10^{17} cm^{-3})	X-center concentration	τ_1 (10^{-10} s)	τ_2 (10^{-10} s)	τ_3 (10^{-10} s)	I_1 (%)	I_2 (%)	I_3 (%)
1.14 ± 0.1	low [a]	1.8 ± 0.2	5.01 ± 0.15	9.60 ± 0.20	26 ± 3	60.3 ± 3.0	8.3 ± 1.0
3.2 ± 0.3	high [b]	1.7 ± 0.2	4.56 ± 0.15	9.06 ± 0.20	29 ± 3	48.1 ± 2.4	16.3 ± 1.0
4.1 ± 0.4	low	1.4 ± 0.2	4.37 ± 0.15	9.87 ± 0.20	22 ± 2	60.2 ± 3.0	16.9 ± 1.0
5.2 ± 0.5	high	1.2 ± 0.2	4.29 ± 0.15	9.09 ± 0.20	23 ± 2	54.3 ± 2.7	21.0 ± 1.5
8.8 ± 0.9	low	1.3 ± 0.2	3.93 ± 0.15	9.62 ± 0.30	12 ± 2	54.5 ± 2.7	31.7 ± 1.5

(*a*) Not revealed to the spectrophotometric analysis.
(*b*) Potassium excess of the order of $5 \cdot 10^{17}$ atoms/cm^3.

It can be noted that:

i) The intensity I_3 increases with increasing F-center concentration. This dependence is shown in Fig. 1; comments on the form of the curve drawn through the experimental points will be made below.

ii) As shown in Fig. 2, the decay rate $\Gamma_3 = 1/\tau_3$ is independent of F-center concentration, while $\Gamma_2 = 1/\tau_2$ follows an increasing trend. The shortest mean life τ_1 varies irregularly; it is very likely that this short-lived component is the sum of two unresolved exponentials.

iii) The presence of a stoichiometric excess of potassium in the form of colloidal color center is ineffective so far as concerns the time spectrum distortion at the attained concentration level ($\sim 5 \cdot 10^{17}$ potassium atoms per cm^3).

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(7) A. SMAKULA: *Zeits. Phys.*, **59**, 603 (1930).
(8) C. BUSSOLATI, A. DUPASQUIER and L. ZAPPA: *Nuovo Cimento*, **52** B, 529 (1967).

According to the interpretation proposed for angular-correlation results by HERLACH and HEINRICH, it seems evident that positrons may be captured by F-centers. The system so formed can be pictured as a Ps-atom bound in a vacant negative ion site. Thus the third component that was revealed in the present experiment is to be interpreted as due to the decay of the ortho-state of this system via 2γ pick-off annihilation.

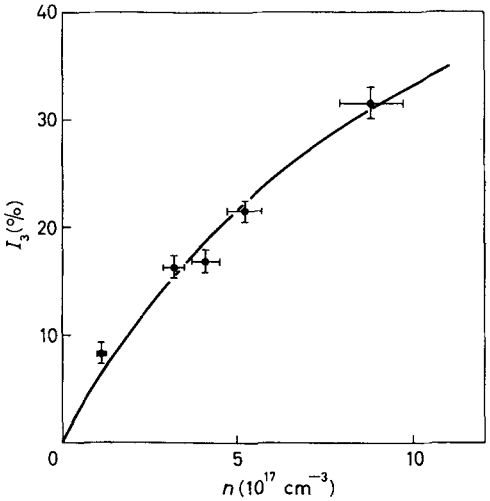

Fig. 1. – Intensity of the third component I_3 as a function of n, number of F-centers per cm^3.

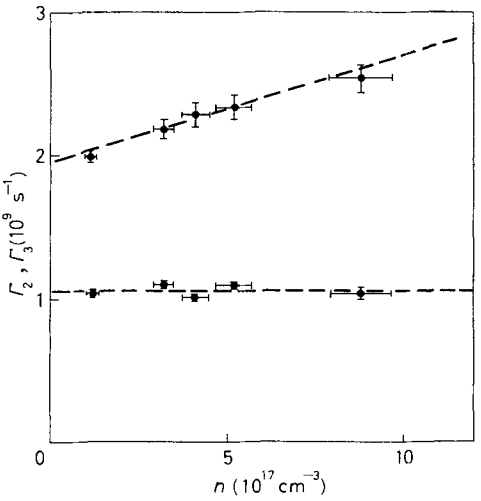

Fig. 2. – Decay rates $\Gamma_3 = 1/\tau_3$ and $\Gamma_2 = 1/\tau_2$ plotted against n, number of F-centers per cm^3.

The ionization probability is fairly low, as indicated by the constancy of Γ_3. The model requires the existence of a para-state component which in the present experiment has probaly not been correctly resolved, as already mentioned. Consistent with the above interpretation, the fitting curve of Fig. 1 has the equation

$$I_3 = \frac{3}{4}\frac{n}{a+n}. \tag{1}$$

In the above formula n indicates the number of F-centers per unit volume and a ($12.39 \cdot 10^{17}$ cm^{-3}) is an empirical constant, calculated by means of a least-squares method. The factor $\frac{3}{4}$ gives the correct asimptotic value, in the hypothesis that ortho and para states are formed in proportion to the respective statistical weights. A mathematical expression like eq. (1) can be derived from the equation system discussed in ref. [3] with the aid of some simplifying assumptions; however, since available experimental information is still scarce, it is safer to consider the drawn curve as an empirical fitting.

The variability of the decay rate Γ_2 indicates that this parameter does not represent a pure annihilation rate, but is the resultant effect of at least two competitive processes: annihilation or, alternatively, transition to a state bound to F-centers.