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Positron Trapping in Indium*

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Abstract. The positron annihilation lineshape was measured in indium in the temperature range between 74 K and the melting point (430 K). A model based on self-trapping of positrons and trapping by vacancies was fitted through the data. A vacancy formation enthalpy of (0.48 ± 0.03) eV was obtained.

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The measurements of the influence on the annihilation parameters of the trapping of positrons by thermally induced vacancies are usually analyzed in terms of the well known two-state trapping model. In most cases the mono-vacancy formation enthalpy H_{1V}^F, obtained in this way agrees reasonably well with the results from other techniques [1]. However for some metals such as the noble metals Cu, Ag and Au the agreement is less satisfactory. This disagreement may be explained with the help of the recently observed anomalous temperature dependence in the sub-vacancy region [2], which was interpreted as an effect of self-trapping of positrons [3]. Based on this self-trapping model, Seeger [3] has shown that for certain metals the mono-vacancy formation enthalpy and entropy can be brought into the more established range of values.

We measured the temperature dependence of the positron annihilation lineshape factor in indium between 74 K and the melting point (430 K). It must be noted that for indium there are no data available from other techniques. Also there is the supplementary problem that the positron measurements are not internally consistent [1]. Angular correlation measurements, as performed by McKee et al. [4], gave for the vacancy formation enthalpy a value of 0.55 eV (± 0.02 eV). This value disagreed with the value of 0.45 eV (± 0.03 eV), as obtained from centroid shift positron lifetime measurements [1]. Triftshäuser [5] investigated the positron trapping in solid and liquid metals and obtained a value of 0.48 eV (± 0.01 eV) for the formation enthalpy in indium by fitting the two-state trapping model to the data, after correction for thermal expansion effects. The data were normalized peak counts and extended from room temperature up to the melting point. Singh et al. [6] obtained a value of 0.39 eV (± 0.04 eV) for H_{1V}^F from a combination of the measurements of mean life and lineshape factor as a function of temperature. However, the set of data extending from room temperature up to 420 K were also analyzed in terms of the simple two-state trapping model without including the effect of the temperature dependence of the positron annihilation parameters in the sub-vacancy region. MacKenzie et al. [7] obtained a value of 0.41 eV from the experimentally observed linear relationship between the threshold temperature and the diffusion coefficient Q. They did not make use of any trapping model calculations but assumed that $Q = 2H_{1V}^F$. We have analyzed the lineshape factor data as a function of temperature in terms of a trapping model, including the effect of self-trapping of positrons.

Experiment

The measurements were done with a 0.14 cc Ge(Li) LEPS coupled to an Ortec model 117 A preamplifier

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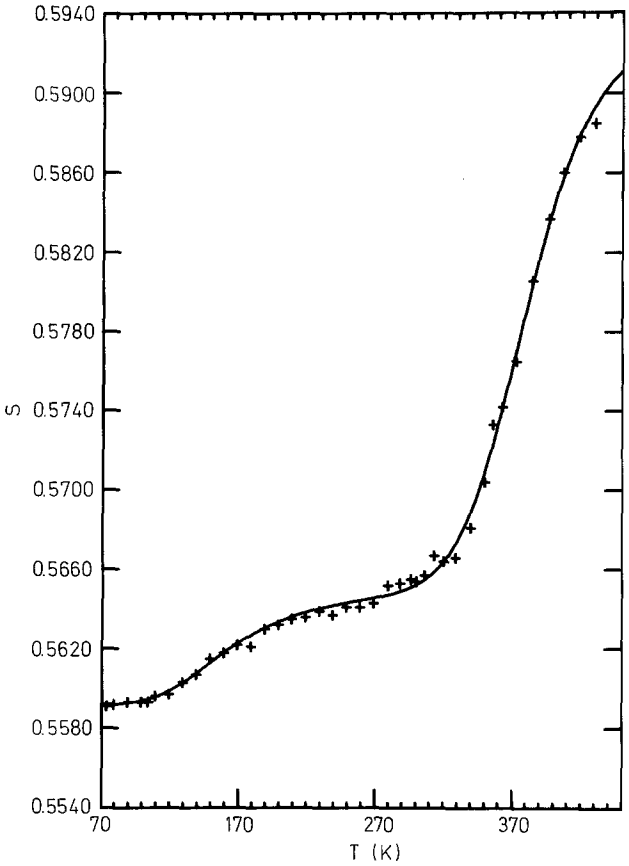

Fig. 1. *S*-factor as a function of temperature in indium. The solid line represents the best fit through the data

with cooled FET. The preamplifier signal was fed into a Tennelec TC205 A amplifier. The analyzer was a Histomat (Intertechnique), with digital zero and gain stabilization. The pulses of two Ortec model 448 precision pulsers were fed into the preamplifier, to obtain two point stabilization. The room temperature was kept constant within 1 C.

The amplifier gain and the conversion gain of the ADC were adjusted to obtain an energy calibration of 0.0427 keV per channel. The resolution of the system was 1.0 keV at the 514 keV gamma line of ^{85}Sr. The peak position stability was better than 0.02% over a period of 1 month.

The samples were 1 cm diameter by 0.25 cm thick disks of 99.9999% pure indium (Koch & Light). As source we used carrier-free ^{22}Na evaporated on 0.1 mil thick mylar foil, covered with a similar foil. Due to the low efficiency of the Ge(Li) LEPS detector each measurement had a duration of 5 hrs, in order to obtain some 7.10^5 accumulated counts.

For the measurements between room temperature and the melting point (429.76 K) the assembly of samples and source was placed in an Elscint model MF-2B furnace in which the temperature was kept constant within 0.5 K. The vacuum was 1.10^{-5} mm Hg. For the low temperature measurements the sample-source sandwich was mounted in an Oxford Instruments CF-100 cryostat, where the temperature could be varied between room temperature and liquid nitrogen temperature, and kept stable within 0.1 K.

The peak position of the 511 keV annihilation line was determined by fitting a Gaussian to the upper 30% of the peak, with the help of a PDP 15/20 computer. Background was carefully subtracted and a *S*-factor was defined in the way, as described by MacKenzie et al. [8]. The central integration was taken over 41 channels for a 511 keV annihilation line with a FWHM of some 60 channels.

The *S*-factor as a function of temperature is illustrated in Fig. 1. Each point is a mean of at least three measurements. The RMS is always less than 0.0009. The *S*-factor at room temperature was measured i) with the sample in the furnace, both at the beginning and at the end of the high temperature cycle ii) with the sample in the cryostat, both at the beginning and at the end of the low temperature cycle iii) with the free sample. All these measurements gave the same value within the statistical accuracy; the mean value was used in the calculations.

Discussion

From Fig. 1 it is clear that below the vacancy trapping threshold at approximately 330 K, the temperature dependence of the lineshape factor is not linear. At the low temperature end the lineshape factor levels off at approximately 100 K to the value $S_f = 0.5592$.

The behaviour of the lineshape factor as a function of temperature is quite similar to that in the case of Cd [2, 3], where the effect in the intermediate temperature region is interpreted in terms of metastable positron self-trapping. In this intermediate temperature region, where no vacancy trapping occurs, the observed lineshape factor S_{nt} is a weighted average of the lineshape factor of completely free positrons (S_f) and the lineshape factor of self-trapped positrons (S_{st}) i.e. [9]

$$S_{nt}(T) = [S_f \lambda_f f_f(T) + S_{st} \lambda_{st} f_{st}(T)] / [\lambda_f f_f(T) + \lambda_{st} f_{st}(T)], \quad (1)$$

where $f_f(T)$ and $f_{st}(T)$ are the fractions of positrons in the free and in the self-trapped state. The expressions

for these fractions can be obtained from statistical considerations and are given by [3]

$$f_f(T) = \{1 + AT^{-3/2} \exp[-\varepsilon(\kappa_0)/kT]\}^{-1}$$
$$f_{st}(T) = 1 - f_f(T) \quad (2)$$

with A and $\varepsilon(\kappa_0)$ constants as defined in [3]. For low temperature $f_{st}(T)$ equals zero and a levelling-off of the S-factor as function of temperature is seen where S reaches the value S_f. In the case where only self-trapping occurs, such a levelling-off should also be seen at the high temperature end, where S_{nt} reaches the value S_{st}. In the actual case, we only can see the levelling off at the low temperature end. At the high temperature end, the saturation effect due to self-trapping is masked by the effect of vacancy trapping.

In order to describe the measured temperature dependence of the lineshape factor, including self-trapping as well as vacancy trapping, we have to consider a trapping model, in which the annihilation characteristics of the "not-trapped" positron are a weighted average of the annihilation characteristics of the completely free positron and the self-trapped positron. The temperature dependence is introduced by i) the temperature dependence of the fraction of free and self-trapped positrons ii) the number of positrons trapping centers (i.e. the vacancy concentration as a function of temperature) and iii) the temperature dependence of the trapping probability. This trapping model has been fully described in [9]. As a result for the lineshape factor as a function of temperature we obtain

$$S = \frac{\lambda_f f_f S_f + \lambda_{st} f_{st} S_{st} + \mu_v C_v S_v}{\lambda_f f_f + \lambda_{st} f_{st} + \mu_v C_v}, \quad (3)$$

where μ_v is the trapping probability for a unit vacancy concentration and $C_v(T)$ is the vacancy concentration which reads

$$C_v(T) = B \cdot \exp(-H^F_{1V}/kT).$$

From [10] it follows that there is little difference between the positron lifetimes at $T = 83\,K$ and at room temperature; therefore we assume, as a first approximation that $\lambda_f = \lambda_{st}$ and assign the value $5.56\ 10^9\ s^{-1}$ to it. The experimental value of $S_f = 0.5592$.

First we analyzed the part of the curve in the temperature region below the onset of vacancy trapping. Using a SIMPLEX [11] fitting technique, we obtained values for A and $\varepsilon(\kappa_0)$. These values were used as starting values to fit the complete curve with expression (3). As a first approximation we assumed the vacancy trapping probability to be temperature independent.

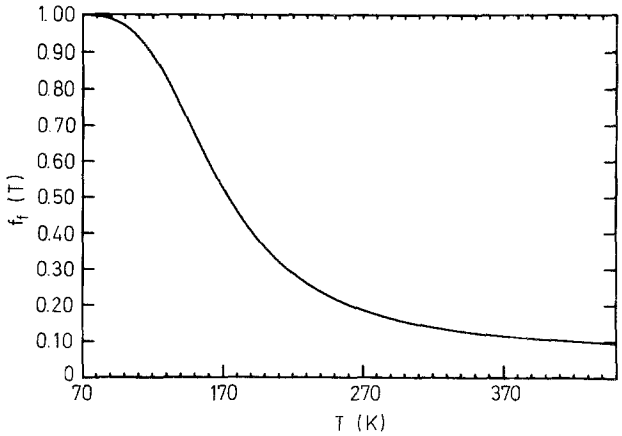

Fig. 2. Fraction of initially free positrons as a function of temperature

Table 1

H^F_{1V} [eV]	References
0.55 ± 0.02	McKee [4]
0.45 ± 0.03	McKee quoted in [1]
0.48 ± 0.01	Triftshäuser [5]
0.39 ± 0.04	Singh [6]
0.41	McKenzie [7]
0.48 ± 0.03	this work

An improvement of the fit could be obtained by taking S_v as a linearly decreasing function of temperature; the physical justification for this has been explained in [9]. Taking λ_f different from λ_{st} [10] did not improve the fit and did not alter the results. A fit with a temperature dependent vacancy trapping probability was not tried, because [9] reported that such a procedure did not alter the goodness of fit. As a final result for the unconstrained fitting of the complete curve we obtain

$$A = (0.91 \pm 0.06)\ 10^6$$
$$\varepsilon(\kappa_0) = (0.090 \pm 0.002)\ \text{eV}$$
$$\mu_v B = (1.6 \pm 0.4)\ 10^{16}\ s^{-1}$$
$$H^F_{1v} = (0.48 \pm 0.03)\ \text{eV}$$
$$S_v = 0.5978 - 0.84\ 10^{-5} \cdot T$$
$$S_{st} = 0.5658 \pm 0.0002$$
$$S_f = 0.5592 \pm 0.0005.$$

The fitted curve is represented in Fig. 1 as a solid line through the data points. In Fig. 2 we have represented the fraction of free positrons as a function of temperature.

If one fits the data above 296 K with a simple two state trapping model with temperature independent trapping efficiency and without applying any correction to the data, then the result is quite different. We obtained $H_{1V}^F = (0.56 \pm 0.04)$ eV and $\mu_v B = (1.76 \pm 0.4)\ 10^{17}\ s^{-1}$, which closely resembles the values obtained by McKee et al. [4]. As pointed out by Seeger [1], a value of 0.56 eV for H_{1V}^F is certainly incompatible with the value of (0.81 ± 0.015)eV [12, 13] for the activation energy of self-diffusion. Table 1 summarizes the vacancy formation enthalpy values obtained by positron annihilation techniques. It would be very desirable to have more experimental data about self-diffusion in indium and vacancy formation enthalpy values measured with techniques other than positron annihilation.

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References

1. A. Seeger: J. Phys. F (Metal Phys.) **3**, 248 (1973)
2. P.C. Lichtenberger, C.W. Schulte, I.K. MacKenzie: Appl. Phys. **6**, 305 (1975)
3. A. Seeger: Appl. Phys. **7**, 85 (1975)
4. B.T.A. McKee, W. Triftshäuser, A.T. Stewart: Phys. Rev. Lett. **28**, 358 (1972)
5. W. Triftshäuser: Phys. Rev. B **12**, 4634 (1975)
6. K.P. Singh, G.S. Goodbody, R.N. West: Phys. Lett. **55A**, 237 (1975)
7. I.K. MacKenzie, P.C. Lichtenberger: Appl. Phys. **9**, 331 (1976)
8. I.K. MacKenzie, J.A. Eady, R.R. Gingerich: Phys. Lett. **33A**, 279 (1970)
9. D. Herlach, H. Stoll, W. Trost, H. Metz, T.E. Jackman, K. Maier, H.E. Schaefer, A. Seeger: Appl. Phys. **12**, 59 (1977)
10. V.H.C. Crisp, D.G. Lock, R.N. West: J. Phys. F (Metal Phys.) **4**, 830 (1974)
11. C. Dauwe, M. Dorikens, L. Dorikens-Vanpraet: Appl. Phys. **5**, 45 (1974)
12. J.E. Dickey: Acta Met. **7**, 350 (1959)
13. G.W. Powell, J.D. Braun: Trans. AIME **230**, 694 (1960)