Appl. Phys. 6, 273—275 (1975)

Applied Physics

Positron Lifetimes in Quenched Al + 0.09 at.-% Mn Crystals

L. J. Cheng
Physics Department*, New York University, New York, N.Y. 10003, USA and Physics Department**, State University of New York at Albany Albany, N.Y. 12222, USA

M. L. Swanson
Atomic Energy of Canada Ltd, Chalk River Nuclear Laboratories, Chalk River, Ontario, Canada

Received 24 September 1974/Accepted 5 November 1974

Abstract. Measurements of positron lifetimes in quenched aluminum + 0.09 at.-% Mn single crystals reveal the existence of a long-lived component whose lifetime and intensity increases and decreases with increasing temperature, respectively. The component is attributed to the annihilation of positrons at voids in the sample formed during quenching.

Index Headings: Positron lifetimes — Defects in solids

Positron annihilation techniques have been useful tools for studying vacancy defects in various solids [1, 2] because a thermalized positron in a solid can be trapped into a vacancy defect where the positron has unique annihilation characteristics. In this short communication, some results from a study of positron lifetimes in quenched aluminum + 0.09 at.-% Mn single crystals are reported.

Single crystal discs of 1.4 cm diameter and 0.1 cm thickness were heated to 600°C in air and quenched into water at room temperature. The samples were kept at room temperature for different times before measurement. The positron lifetimes in these quenched samples were measured at temperatures of 77–300 K by using a conventional positron lifetime spectrometer which had a full width of 300 psec at half maximum for a ^{60}Co source at the setting for the positron lifetime measurements. A Na^{22} positron source was sandwiched by two crystals. By using a computer analysis to fit the data with several exponential decay curves, we have found that each delay time spectrum fits best with two components plus a constant background. The two components correspond to two positron lifetimes. The short component has a lifetime of 190 psec which does not have any significant temperature dependence, but does vary slightly from one quenching run to the other. This short component can be attributed to the average lifetime of positrons annihilating with electrons in the perfect part of the crystal and in small vacancy defects introduced during quenching. However, the long component has significant temperature dependence in its lifetime and intensity, as shown in Fig. 1, where τ_2 is the lifetime and I_2 is the intensity. Both the lifetime and the intensity vary from one quenching run to the other, however, the appearance of the temperature dependence for all the runs is the same, as shown in the figure. A 30 min annealing at 150°C had no effect on the long component, but a 30 min annealing at 350°C made it disappear almost completely. A weak component of 1 – 2% with an even longer lifetime (> 500 psec) could be seen through computer analysis of two

* Supported by the National Science Foundation of USA.
** Present address and supported by the Office of Naval Research of USA under contract N 00014-90-0296.

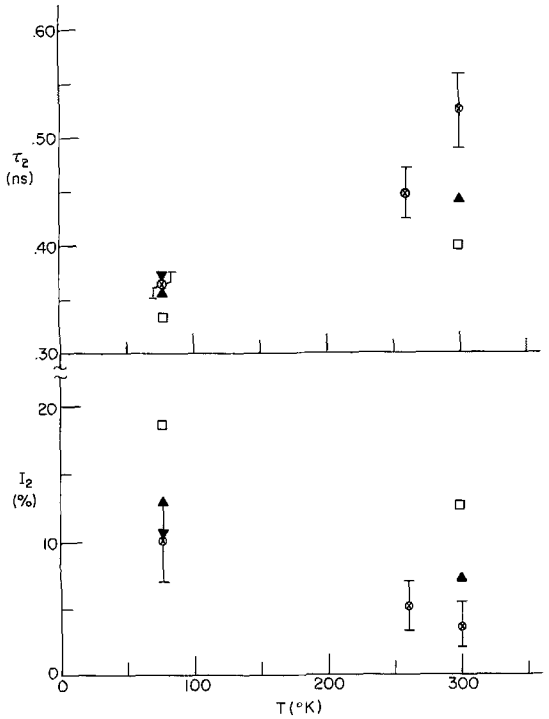

Fig. 1. Lifetime and intensity of the long component versus temperature. Different symbols represent different runs

component fitting after the 350°C annealing, however its temperature dependence is different from the one before annealing. From the above observations, it is clear that the long component shown in Fig. 1 is due to the presence of quenched defects.

Voids have been observed in aluminum quenched from 600°C [3] and their annealing temperature was reported to be around 200–350°C [4]. The equilibrium vacancy concentration in Al at 600°C is about $10^{18}/\text{cm}^3$ which is enough for forming voids with a concentration detectable by the positron technique. Possibly, the presence of Mn helps the nucleation of voids. Cotterill *et al.* [5] reported an observation of long positron lifetimes of 467 – 595 psec in neutron irradiated Mo which were correlated with voids in the sample. The lifetime of the long component observed at 300 K in the present study is similar to their values. It has also been shown that the angular correlation of positron annihilation in Al is markedly affected by voids which are produced by neutron irradiation [6]. The above mentioned observations support the thought that the defects responsible for the observation of the longer component in the present study are voids. It should be noticed that the positron technique is sensitive to small voids which is not normally observable under electron microscopy. What is the lower limit in size of voids in metals? This is still an open question.

The temperature dependence of the long component which we observed is interesting. By using the trapping model of Seeger [2] first proposed by Brandt [7], we have estimated the positron trapping rate of the defect responsible for the long component. In this model, we have considered trapping by two types of defects, voids and small defects, with respective trapping rate and concentration; σ_v, C_v and σ_d, C_d. The nature of the small defects is not known, presumably they are vacancy-type defects and, possibly, dislocations. The positron lifetime at the small defects, τ_d, is only slightly larger than that in the perfect part of crystal, τ_c. It is assumed that all positrons are initially free in the perfect part of crystal and that detrapping is negligible. The short lifetime τ_1 observed in our study is the average lifetime due to annihilation of positrons in the perfect part of the crystal and at small defects. Then, the intensity of the long lifetime component, caused by annihilation at voids, is derived to be

$$I_2 = \frac{\sigma_v C_v}{1/\tau_c + \sigma_d C_d + \sigma_v C_v - 1/\tau_2} \tag{1}$$

and the short lifetime

$$\tau_1 = \tau_d \frac{(1/\tau_d + \sigma_d C_d)(1/\tau_c + \sigma_v C_v + \sigma_d C_d - 1/\tau_2)}{(1/\tau_c + \sigma_d C_d + \sigma_v C_v)(1/\tau_2 + \sigma_d C_d - 1/\tau_2)} \tag{2}$$

where τ_2 is the positron lifetime at voids. Taking $1/\tau_c + \sigma_d C_d \gg 1/\tau_2$, we have

$$\tau_1 = \tau_c \frac{(1 + \tau_d \sigma_d C_d)}{(1 + \tau_c \sigma_d C_d)}. \tag{3}$$

It is known that the positron lifetime in a well-annealed pure Al crystal is about 170 psec, which varies slightly with temperature, due to thermal expansion. Based on the known thermal expansion coefficient of Al, it is estimated that τ_c increases about 2% when temperature rises from 70 K to 300 K. $\sigma_d C_d$ is estimated to be $2.3 \times 10^9/\text{sec}$ from (3) with the observed value of 190 psec for τ_1 and the known positron lifetime of 240 psec at vacancies in Al for τ_d. Then, from the observed temperature dependence of I_2, we have found that σ_v varies as $T^{-\alpha}$ with $0.3 < \alpha < 0.8$. It has been noticed that this result is relatively insensitive to a variation in σ_d.

Our result that the trapping rate increases with decreasing temperature in the range from 77 K to

300 K is consistent with the result of Connors *et al.* [8] in quenched Cd and the theoretical prediction of Seeger [9]. However, the present result differs from the work of McKee *et al.* [10] who observed temperature independence of positron trapping by vacancies in quenched gold. The difference can be explained by means of the difference in sample nature, defect type, and possibly experimental technique. However, our observation of no significant temperature dependence of the short lifetime component is consistent with the result of McKee *et al.* Simple calculations, based on (2) or (3), have shown that less than 3% increase of τ_1 can be expected when temperature rises from 77 K to 300 K, if either σ_v or σ_d or both vary with $T^{-1/2}$ and τ_c increases with thermal expansion.

It was also noticed that the error in our data could be as great as 3%. Using a new computer analysis on their data of the temperature dependence of the positron lifetime for studying vacancy formation in pure aluminum, pure gold, and an aluminum alloy with 1.7 at.-% Zn at high temperatures, Hall *et al.* [11] found that the fit to their data was significantly improved by assuming a temperature dependence of the positron trapping rate of $T^{1.2\pm0.3}$ for Al, $T^{0.5\pm0.2}$ for Au, and $T^{1.1\pm0.2}$ for the Al alloy. It appears that the data available now on the temperature dependence of the positron trapping rate at vacancy defects in metals are controversial. Because of the diffuse nature of the positron wave function, it is possible that the quantum transition rate approach applies to positron annihilation at a high concentration of small defect (vacancies), whereas the mobility-trapping rate approach [2, 9] applies for annihilation at a lower concentration of larger defects (voids). The former theory leads to a temperature-independent trapping rate, while the latter leads to a $T^{-1/2}$ dependence.

Calculations of Hodges and Stott [12] indicate that positron bound states at voids in metals are localized at surfaces. Their corrected estimate for the lifetime at voids in Al is ~ 600 psec, in approximate agreement with our value measured at 300 K. Recently, Nieminen and Manninen [13] did numerical estimates for binding energies, annihilation lifetimes, and 2γ-angular correlation curves for the positron surface state in aluminum using three different model potentials. Their values agree well with the measured lifetime at 77 K. The increase of τ_2 with temperature observed in the present study is consistent with their model in which the potential at the surface is a deep potential "trough" with a vertical increase at the metal surface and a slow increase toward the center of the void. It is possible that this kind of potential can make the trapped positron at a higher (thermally excited) bound state spend less time in the metal than that at a lower state. By a rough argument, the lifetime is inversely proportional to the time spent by the positron in the metal. Then, the lifetime can increase with temperature as observed in the present study. Our observation shows that the temperature dependence of the positron lifetime at a defect can be measured which can certainly provide further information important to our understanding of defects in crystalline solids.

Acknowledgement. One of the authors (L.J.C.) would like to acknowledge the hospitality of W. Brandt and his laboratory where the measurements of positron lifetime were done.
The authors wish to thank J. W. Corbett, B. T. A. McKee, and P. Sen for helpful discussions.

References

1. For example, L.J.Cheng, C.K.Yeh, S.I.Ma, C.S.Su: Phys. Rev. B**8**, 2880 (1973)
2. A.Seeger: J. Phys. F. (Metal Phys.) **3**, 248 (1973)
3. M.Kiritani, Y.Shimomura, S.Yoshida: J. Phys. Soc. Japan **19**, 1624 (1964)
4. J.Jostsons, E.L.Long, Jr., J.O.Stiegler, K.Farrell, D.N.Braski: In: *Radiation-Induced Voids in Metals*, Ed. by J.W.Corbett and L.C.Ianniello (U.S. Atomic Energy Commission, Oak Ridge, Tennessee 1972) p. 363
5. R.M.J.Cotterill, I.K.Mackenzie, L. Smedskjaer, G.Trumpy, J.H.O.L.Träff: Nature **239**, 99 (1972)
6. W.Triftshauser, J.M.McGervey, R.W.Hendricks: Phys. Rev. B**9**, 3321 (1974)
7. W.Brandt: In: *Positron Annihilation*, Ed. by A.T.Stewart, L.O.Roellig (Academic Press, New York 1967) p. 155
8. D.C.Connors, V.H.C.Crisp, R.N.West: Phys. Letters **33**A, 180 (1970);
   D.C.Connors, J.C.Bowler: Phys. Letters **43**A, 395 (1973)
9. A.Seeger: Phys. Letters **40**A, 135 (1972); ditto **41**A, 267 (1972)
10. B.T.A.McKee, H.C.Jamieson, A.T.Stewart: Phys. Rev. Letters **31**, 634 (1973)
11. T.M.Hall, A.N.Goland, C.L.Snead, Jr.: BNL report 18771 (1974) and to be published in Phys. Rev.
12. C.H.Hodges, M.J.Stott: Solid State Commun. **12**, 1153 (1973)
13. R.Nieminen, M.Manninen: Solid State Commun. **15**, 403 (1974)