Elemental melts studied by positron lifetime and Doppler broadening measurements

1989 J. Phys.: Condens. Matter 1 SA97

(http://iopscience.iop.org/0953-8984/1/SA/014)

J. Phys.: Condens. Matter 1 (1989) SA97–SA106. Printed in the UK

Elemental melts studied by positron lifetime and Doppler broadening measurements

H-E Schaefer†, W Eckert†, J Briggmann‡ and W Bauer†
† Institut für Theoretische und Angewandte Physik, Universität Stuttgart, Postfach 801140, 7000 Stuttgart 80, Federal Republic of Germany
‡ Institut für Physik, Max-Planck-Institut für Metallforschung, Postfach 800665, 7000 Stuttgart 80, Federal Republic of Germany

Received 28 February 1989

Abstract. Positron lifetime measurements were performed in Al, In and Ge in the solid and molten phases. In the melts the positrons are found to be annihilated in free volumes the size of a few atomic volumes. These results are discussed together with those of earlier studies on molten Ga, Hg and Na. The influence of the fast fluctuations and that of the presence of the positron on the size of the free volumes in the melt are considered.

1. Introduction

The structure of elemental melts has been studied extensively by x-ray and neutron scattering [1, 2], yielding information on atomic spacings and coordination numbers [1]. On the basis of the shapes of the structure factors $S(q)$ obtained from these scattering experiments three types of molten metals or semiconductors can be distinguished [1]:

(i) Simple metals, like the alkali metals, Al, Pb, and the noble metals, with a symmetric first peak of $S(q)$. These metals show an increase in the specific volume ($\Delta V/V_s > 0$) at the melting transition and retain nearly the same nearest-neighbour distances (V_s designates the specific volume of the solid at the melting temperature T_M). The changes of the coordination numbers in the melt compared with those measured in the crystalline solid are moderate (see table 1 later). The structure factors of these melts are in qualitative agreement with those calculated from the Percus–Yevick [3] solution of the hard-sphere model of a fluid [4] when an appropriate value of the hard-sphere packing fraction η is used.

(ii) Elemental melts like In, Zn, Cd with an asymmetric first peak in $S(q)$. These asymmetries are commonly ascribed to covalent bonding contributions which may be responsible for the non-close-packed solid structure of these elements with lowered crystalline symmetry compared to cubic metals.

(iii) Melts with a shoulder on the high q-side of the first peak in $S(q)$ as is observed for the elemental semiconductors Si and Ge. These elements exhibit a drastic decrease in the specific volume ($\Delta V/V_s < 0$) upon melting. The structure factor $S(q)$ in liquid Ge and its variation with temperature has been discussed [5, 6] in terms of a mixture of covalently bound tetrahedra separated by hard-sphere ions which gives rise to the metallic properties of the melts of semiconductors.

Table 1. Positron lifetimes in solid and liquid elements. Symbols are defined as follows: T_M, melting temperature; $\Delta V/V_s$, change of volume at the solid–liquid phase transition; τ_f and τ_{1V}, positron lifetimes in the free state and in vacancies, respectively; τ^s and τ^l, positron lifetimes at T_M in the solid and liquid states, respectively; r_1^s and r_1^l, nearest-neighbour distances in the solid and liquid, respectively; Z^s and Z^l, coordination numbers in the solid and liquid, respectively. For Ge τ_{1V} after electron irradiation is given [27].

Element	T_M (K)	Crystal structure	τ_f (ps)	τ_{1V} (ps)	τ^l (ps)	$\tau^l - \tau^s$ (ps)	$\Delta V/V_s$ (%)	r_1^s (Å)	r_1^l (Å)	Z^s	Z^l
Al[a]	933	FCC	159	234	243	9	6.5	2.86	2.82[b]	12	11.5[b]
			166[c]	249[c]	260[c]	11[c]					
In[a]	429	tetragonal	197	265	253	−12	2.0	3.24	3.23[b]	4+8	11.6[b]
								3.37			
Ge[a]	1210	diamond	232	279[c]	244	12	−5.1	2.44	2.82[b]	4	6.8[b]
Hg[d]	234	rhombohedral	182	(266[e])	261	79	3.7	3.00	3.07[b]	6	10.0[b]
Ga[f,g]	303	orthorhombic	190	(248[e])	260	70	−3.2	2.43	2.82[b]		10.4[b]
								2.79			
Na[h]	371	BCC	338		353	15[f]	2.5	3.71	3.81[b]	8	10.4[b]

[a] Present work
[b] Ref. [1]
[c] Ref. [27]
[d] Ref. [24]
[e] Theoretical values in ref. [28]
[f] Ref. [25]
[g] Ref. [26]
[h] Ref. [29]

No microscopic information on the size of the free volumes in the melt can be deduced directly from the scattering experiments and the above discussion. Hill [7] first suggested that one could derive the probability of free volumes in a melt as a function of their size from the experimentally determined radial atomic distribution function. Davidović and co-workers [8] assumed free volumes of the size of a missing atom to be responsible for the structural and electronic changes characteristic of elemental melts compared with the corresponding solids. According to their results the structure factors $S(q)$ of melts can be understood quantitatively by atomic concentrations of vacancy-like free volumes of between 2×10^{-3} and 5×10^{-2} in the case of metals or between 10^{-5} and 10^{-4} in the case of semiconductors.

Positron annihilation techniques have proved most useful [9–12] in the investigation of vacancy-like defects in solids. In melts of low-melting-point metals the study of the angular correlation of the positron annihilation radiation was employed in early years in order to investigate their electronic structure [13–15]. Later on it became clear that the changes of the angular correlation curves observed in some metals upon melting have to be ascribed to the annihilation of the positrons in free volumes of the melt [16].

Measurements of changes in the angular correlation and the Doppler broadening of the annihilation γ-line upon melting were performed on Cu [17], Ni [18], Pb, In [19], Cd [20], Sn [21], and Bi [22], without yielding detailed information about the size of free volumes in the melts.

Positron lifetime measurements from which specific information on vacancy-type defects in solid metals can be obtained [23], have only been performed on the melts of non-close-packed metals with low melting points [24–26]. In these metals either the nearest-neighbour atomic distance r_1 or the coordination number Z changes upon melting and a decrease of the specific volume may even occur (e.g. Ga).

In this paper results are reported of positron lifetime measurements in the melts of Al, In, and Ge. These experiments were performed in order to study the size of the free volumes where the positrons are annihilated. The sizes estimated from these measurements have to be considered as average values due to the spatial size distribution and fast atomic fluctuations in the melt during the positron lifetime. In addition, the presence of the positron may affect the size of the free volume where the annihilation event occurs. It should be emphasised here that the sizes of the free volumes deduced from positron lifetime measurements can be considered as characteristic of the unperturbed melt *only if* the positron–ion interaction is negligible (see § 3).

In Al, where the nearest-neighbour distance of atoms and the coordination number change only slightly upon melting (see table 1), a reasonable estimate of the size of the free volumes where the positrons are annihilated in the melt can be deduced from a comparison of the positron lifetime in thermal equilibrium vacancies below T_M and its lifetime in the melt. In addition, positron lifetime measurements upon melting of non-close-packed crystals like In with a tetragonal and Ge [11] with a diamond-lattice structure will be presented. Finally some aspects of the positron–ion interaction in the melt and its consequences on the assessment of free volumes in the unperturbed melt will be discussed.

2. Experimental procedures

The positron lifetime measurements on Al [30] discussed here (see figure 1) were performed by means of a fast–slow spectrometer (with a time resolution (FWHM) of 360 ps) after alloying the ^{58}Co positron emitter (3×10^6 Bq) to the Al specimen. The measurements on the Al specimen were performed between 300 and 1258 K in a high-purity graphite container which was sealed in a quartz vial under high vacuum. For the simultaneous Doppler broadening studies a Ge detector with an energy resolution (FWHM) of 1.3 keV at 497 keV was used. The positron lifetimes in In and Ge were measured by means of the $\beta^+\gamma$ technique (with a time resolution (FWHM) of 245 ps and 190 ps, respectively) employing the MeV positron beam at the Stuttgart Pelletron accelerator [31] and electron-beam heating of the specimens in a high-vacuum chamber [31].

High-purity specimens of Al (from VAW, with a residual resistivity ratio of 30000), In (Ventron Alfa Produkte, of purity 99.9999%), and Ge (Czochralski grown, $\rho \geqslant 50\ \Omega$ cm) were used for the present studies.

3. Experimental results and discussion

In *aluminium*, the well known S-shaped temperature variation of the mean positron lifetime [32] is observed in the solid state due to the formation of thermal vacancies (see figure 1). At the solid–liquid transition $\bar{\tau}$ increases by 9 ps and stays constant up to 1258 K. The positron lifetimes determined in the present experiments for solid Al are a little lower than in earlier studies [32]. This may be due to a contribution of 511 keV–511 keV coincidences in the positron lifetime spectra originating from the reduced energy window separation between the prompt 810 keV and the 511 keV annihilation quanta when a ^{58}Co source is used instead of ^{22}Na.

Similar variations of $\bar{\tau}$ with temperature were found in measurements on Al at 296, 873 and 1173 K [27] (see table 1) by means of the $\beta^+\gamma$ coincidence technique. Slightly

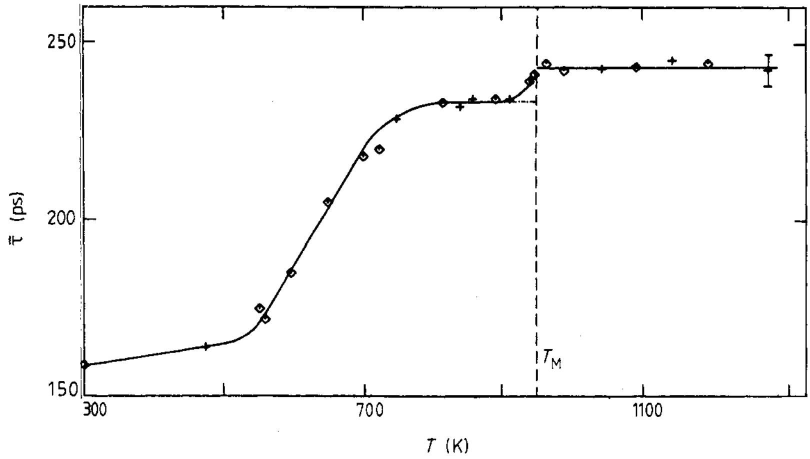

Figure 1. Temperature variation of the mean positron lifetime $\bar{\tau}$ in solid and liquid Al (T_M is the melting point). ◇ and + indicate different runs.

below T_M an increase in $\bar{\tau}$ may occur (see figure 1) which could be due to premelting effects [33] on interfaces or surfaces.

In assessing the change of the positron lifetime upon melting and the size of the free volumes in which the positrons are annihilated in the Al melt, we can consider first the theoretical predictions of positron lifetimes in vacancies in solids. The positron lifetime in a divacancy in crystalline Al has been calculated [23] to be 20 ps higher than in a monovacancy. Taking into account the uncertainty limits of this value arising from the assumptions made in the calculations, the increase of $\bar{\tau}$ upon melting in Al may be ascribed to positron annihilation in free volumes in the melt with a size of about 1 to 3 atomic volumes. This is in qualitative agreement with the findings of an *R*-parameter analysis of angular correlation studies in solid and liquid Al [19]. The results obtained in [19] indicate that in molten Al the free volumes where the positrons are annihilated are a little larger than single lattice vacancies, but much smaller than voids induced by neutron irradiation in an Al crystal.

Longer positron lifetimes characteristic of a larger size of free volumes in the melt were not observed. This may be due to rapid atomic fluctuations [34] which give rise to the high self-diffusivities in melts [4, 35]. These fluctuations occur on a time scale of 10^{-12}–10^{-11} s, much shorter than the positron lifetime, so that the sizes of the free volumes probed by the positron fluctuate during this lifetime. The sizes of free volumes in melts are expected to be distributed between values of less than an atomic volume, as detected in amorphous metals [36], and of several atomic volumes. Therefore, the positron lifetimes reported here for the melts of Al and other elements, as well as the sizes of free volume attributed to these lifetimes, have to be considered as average values. A detailed interpretation of the measured lifetimes would require a statistical treatment of the various environments of the positron in the melt during its lifetime and a calculation of the corresponding annihilation rates. Moreover, the possibility that the presence of the positron may influence the sizes of the free volumes in the melt [37, 38] has to be taken into account. This will be discussed below.

No variation of $\bar{\tau}$ is observed in the Al melt between T_M and 1258 K, although the specific volume of the melt increases [39] with temperature. This increase may originate

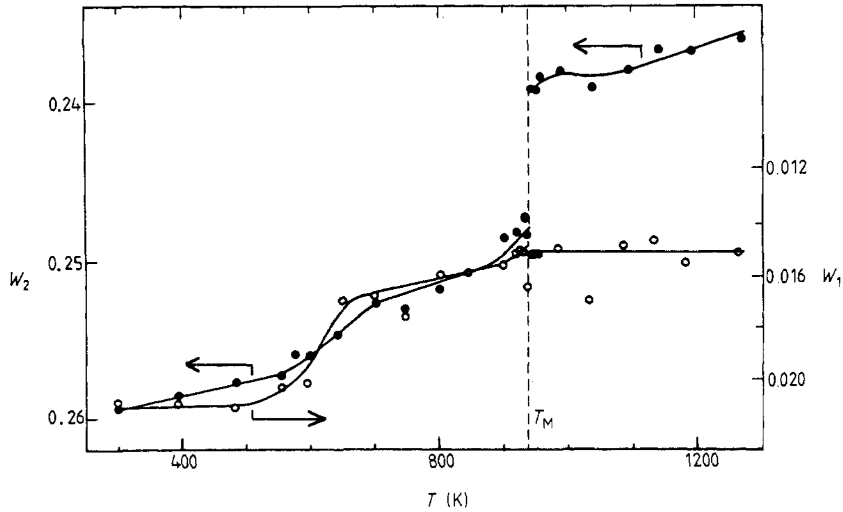

Figure 2. Temperature variations of the Doppler broadening 'wing' parameters. ○, W_1; ●, W_2. For definitions of W_1 and W_2 see figure 3.

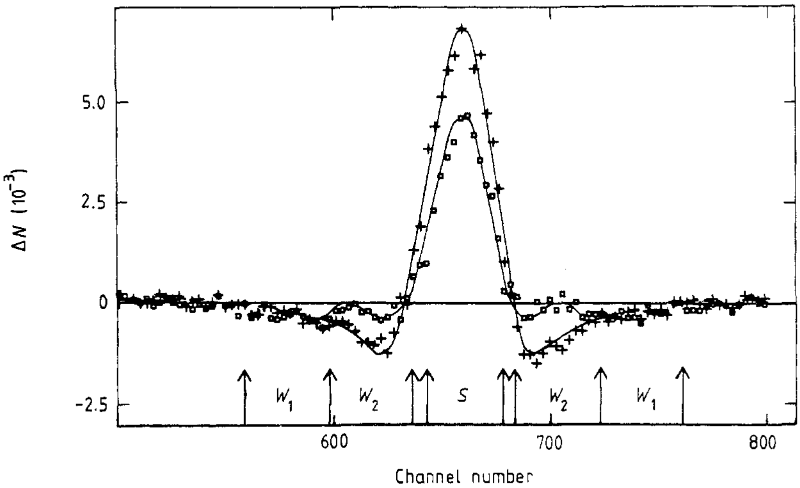

Figure 3. Difference curves of Doppler broadening spectra measured at 830 K (vacancy trapping) and 394 K (e^+ 'free' state), designated by □; measured at 1177 K (melt) and 394 K, designated by +. The summation areas for the 'wing' parameters W_1 and W_2 (plotted in figure 2) and for the peak parameter S are indicated.

from an increase in the number density of free volumes, which remains undetected in the present experiments due to saturation trapping of positrons.

The temperature variations of the Doppler broadening parameters W_1 and W_2 (for a definition of which see figure 3), measured simultaneously with the positron lifetime, show a surprising behaviour at T_M as demonstrated in figure 2. The wing parameter W_1, which is characteristic of e^+ annihilations with core electrons, exhibits only a small increase at T_M, this being considered to be characteristic of an increase of the free volume of a trap in a similar way as the change in $\bar{\tau}$ (figure 1). In contrast, W_2 (and similarly the central S-parameter [30]), which is characterised by the momenta of electrons at the Fermi edge, shows a strong increase at T_M. This behaviour may be ascribed to a stronger positron localisation and therefore a different contribution of the positron to the e^+e^-

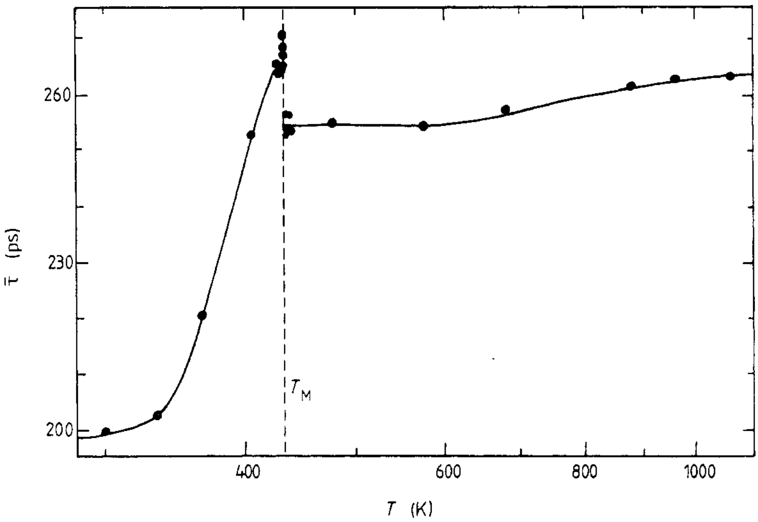

Figure 4. Temperature variation of the mean positron lifetime $\bar{\tau}$ measured in solid and liquid In. Note the logarithmic temperature scale.

momentum at the Fermi edge in a thermal vacancy compared with that in free volumes in the melt. This will be discussed by making use of difference curves of the Doppler broadening data. The different momentum distributions of the e^+e^- pairs in thermally formed vacancies (at 838 K) and in the melt (at 1177 K) are shown in figure 3 by the corresponding difference curves with respect to the 'free' delocalised state (at 394 K). The positron localisation in the vacancies gives rise to maxima in the difference curve at the Fermi edge which are covered by the W_2 range (open squares in figure 3). In the melt (crosses in figure 3) these maxima vanish, indicating a reduction of the positron localisation as compared with that in a lattice vacancy.

In the positron lifetime measurements on *indium* (see figure 4), the present $\beta^+\gamma$ measurements show quantitatively the same S-curve due to thermal vacancy formation below T_M as was observed in earlier $\gamma\gamma$ studies [40]. An additional small increase in $\bar{\tau}$ is suspected to appear just below T_M, similar to the situation in Al. This increase is, however, much smaller than reported for impure In just below T_M [26]. In contrast to the results for Al, a decrease in $\bar{\tau}$ by about 12 ps is observed at the solid–liquid transition in In. This may be attributed to positron annihilation in the In melt in free volumes of a size smaller than that of lattice vacancies. In the In melt the free volume of a missing atom is expected to be reduced compared with that of a lattice vacancy due to the increased atomic packing density. This increase is indicated by an increase in the atomic coordination number and is also shown by the fact that the increase in the specific volume $\Delta V/V_s$ is substantially smaller than in close-packed metals (see table 1). From this we conclude that in the In melt the positrons are on average annihilated in free volumes of the size of at least one missing atom.

In positron age–momentum measurements in molten In [26] and positron lifetime measurements in the Al melt [30], only one type of annihilation characteristic and one lifetime component can respectively be detected within the present experimental uncertainties. From this we may deduce that all positrons have a rather similar history with respect to the sizes of the free volumes they encounter during their lives. More detailed information on the distribution of free volumes may be expected from measurements with improved statistics.

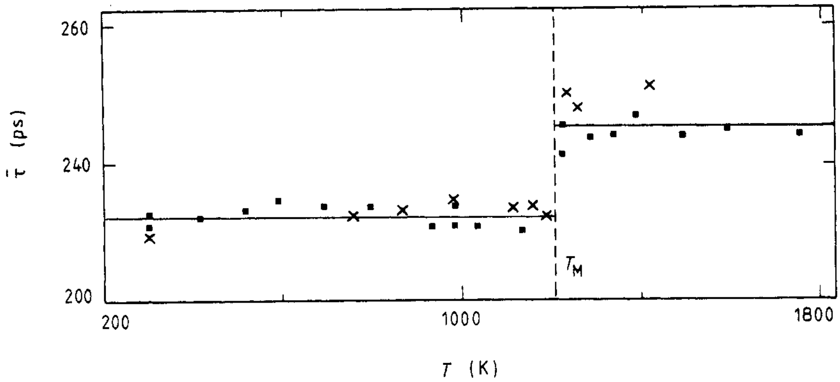

Figure 5. Temperature variation of the mean positron lifetime $\bar{\tau}$ measured in solid and liquid Ge. ■ and × indicate two different runs.

Between 580 and 1060 K the positron lifetime in molten In increases by about 8 ps. This has to be ascribed to a size increase of the free volumes probed by the positrons.

At the melting transition in *germanium* the atomic packing density increases, as demonstrated by the increases in the relative mass density $\Delta\rho/\rho_s$ $(-\Delta V/V_s)$ and coordination number Z (see table 1). Nevertheless, the positron lifetime increases at T_M (see figure 5) which again points to the annihilation of the positrons in free volumes. In assessing the size of these free volumes we have to bear in mind the following issues:

(i) in solid Ge no high-temperature thermal vacancies can be detected by positron annihilation techniques [11, 27]. The positron lifetime of 245 ps observed in the melt could be compared to the value $\tau_V = 279$ ps of radiation-induced vacancies [11, 27]. However, a direct comparison is not very useful for a size estimate of the free volumes in the melt on an atomic scale because of the strong change in the atomic packing upon melting.

(ii) Moreover, Ge assumes metallic properties in the melt with 'enhanced' e^+e^- Coulomb correlation compared with semiconductors. This leads to an increased annihilation rate, so that the positron lifetime measured in the melt is expected to be characteristic of a larger free volume than the same lifetime in the semiconducting solid.

We therefore think that in molten Ge the positrons are again annihilated in free volumes of the size of one or a few missing atoms. Between T_M and 1760 K a constant value of $\bar{\tau}$ is observed within experimental uncertainty (figure 5) although slight structural changes in the melt may be derived from the neutron scattering data of Gabathuler and Steeb [5].

At this point the results of positron lifetime measurements on other metallic melts available in the literature should be included in the discussion. In metals without thermal vacancy formation in the solid (e.g. Ga, Hg), $\bar{\tau}$ strongly increases at the melting transition beyond the value calculated for lattice vacancies (see table 1) although in the case of Ga the specific volume is reduced in the melt. This again indicates that positron annihilation in the melt occurs in free volumes of a few missing atoms. It should be pointed out that Kishimoto and Tanigawa [26] observed the existence of two different annihilation states in liquid Ga by positron age–momentum studies, in which coincident measurements of positron lifetime and Doppler broadening of the annihilation γ line were performed. These authors postulate atom clusters with a similar structure as the solid and a high electron density as positron annihilation sites in the melt in addition to free volumes.

They atrributed the decrease in $\bar{\tau}$ with increasing temperature above T_M to a decrease in the size and an increase in the number of clusters. From this type of study further details of positron annihilation in melts may be obtained.

In alkali metals like Na, monovacancies are formed thermally in measurable concentrations below T_M [41] without giving rise to a significant increase in $\bar{\tau}$ [29]. In Na the positron lifetime, which is much higher in the free state than in the other elements discussed here, increases at the melting transition (see table 1). This further indicates the presence of free volumes of a few missing atoms at the annihilation site.

The presence of the positron and its interaction with the surrounding ion cores may be important in interpreting the positron lifetime data in melts in terms of the sizes of free volumes. This interaction is also to be expected for a positron in a lattice vacancy, but it has not been taken into consideration in theoretical studies of the positron lifetime [23, 42]. For the positron–ion interaction in the melt two cases can be imagined:

(i) the positron is strongly localised in the free volumes of the melt, as would be expected for lattice vacancies in metals [23]. Then it may repel the neighbouring positive ion cores, which are highly mobile, and selectively stabilise a free volume of one or a few missing atoms during its lifetime. The positron lifetime measured in the melt is then considered to characterise a 'bound' state of the positron in a free volume which is determined by the positron–ion interaction. This free volume, which may migrate through the melt as a result of the rapid atomic fluctuations is thought not to be representative for the unperturbed melt. In this picture the increase of the positron lifetime in the In melt at high temperatures (see figure 4) may be due to the formation of free volumes that are on average larger than those of the 'bound' state.

(ii) The localisation of the positron in a free volume in the melt is substantially reduced compared with the situation in vacancies in metal crystals. This appears to be supported by the present results for the Doppler broadening in molten and solid Al (see figure 3). In this case the repulsive interaction between the positron and the ion cores neighbouring the free volume may be reduced or negligible. Then the measured positron lifetime may characterise the distribution of sizes of free volumes in the melt without affecting them. The high-temperature increase of $\bar{\tau}$ in the In melt may then be attributed to a general increase in the sizes of the free volumes in the melt.

Greater understanding of the annihilation of positrons in elemental melts and their interaction with free volumes may be attained by measurements on molten elements with various atomic masses, by applying the positron age–momentum technique, pressure experiments, etc.

4. Summary

From the results of positron lifetime studies presently available, we conclude that in elemental melts near T_M positrons are predominantly annihilated in free volumes of the size of one or a few missing atoms.

To obtain a more detailed interpretation of the positron lifetimes the spatial distribution of the sizes of these free volumes and their fluctuations with time should be taken into account. The sizes of free volumes deduced from positron lifetime measurements can be considered as representative for the unperturbed melt *only* if the positron–ion interaction is negligible.

Acknowledgments

The authors appreciate the continuing interest of Professor A Seeger in this work. They are indebted to the staff of the Stuttgart Pelletron accelerator. Helpful discussions with Professors S Steeb and F Noack, R Würschum and Dr S Bending are gratefully acknowledged. The present studies were supported financially by the Deutsche Forschungsgemeinschaft.

References

[1] Waseda Y 1980 *The Structure of Noncrystalline Materials* (New York: McGraw Hill)
[2] March N H, Street R A and Tosi M 1985 *Amorphous Solids and the Liquid State* (New York: Plenum Press)
[3] Percus J K and Yevick G J 1958 *Phys. Rev.* **110** 1
[4] Shimoji M 1977 *Liquid Metals* (London: Academic Press)
[5] Gabathuler J P and Steeb S 1979 *Z. Naturf.* a **34** 1314
[6] Popescu M 1981 *J. Physique Coll.* **42** C4 1043
[7] Hill T L 1958 *J. Chem. Phys.* **28** 1179
[8] Davidović M, Stojic M and Jović Dj 1984 *J. Non-Cryst. Solids* **61–2** 517
[9] 1979 *Positrons in Solids* ed. P Hautojärvi (Berlin: Springer)
[10] Schaefer H-E 1987 *Phys. Status Solidi* a **102** 47
[11] Würschum R, Bauer W, Maier K, Major J, Seeger A, Stoll H, Carstanjen H-D, Decker W, Diehl J and Schaefer H-E 1989 *Positron Annihilation* ed. L Dorikens-Vanpraet, M Dorikens and D Segers (Singapore: World Scientific) p 671
[12] Forster M, Mundy J N and Schaefer H-E 1989 *Positron Annihilation* ed. L Dorikens-Vanpraet, M Dorikens and D Segers (Singapore: World Scientific) p 833
[13] Gustafson D R and Mackintosh A R 1963 *Phys. Lett.* **5** 234
[14] Kusmiss J H and Stewart A T 1967 *Adv. Phys.* **16** 471
[15] West R N, Borland R E, Cooper J R A and Cusack N E 1967 *Proc. Phys. Soc.* **92** 195
[16] Itoh F 1976 *J. Phys. Soc. Japan* **41** 824
[17] Nanao S, Kuribayashi K, Tanigawa S and Doyama M 1977 *J. Phys. F: Met. Phys.* **7** 1403
[18] Fluss M J, Smedskjaer L C, Chakraborty B and Chason M K 1984 *J. Phys. F: Met. Phys.* **13** 817
[19] Triftshäuser W 1975 *Phys. Rev.* B **12** 4634
[20] Rudzinska W, Szymanski C Z, Chabik S and Rozenfeld B 1986 private communication
[21] Rudzinska W, Szymanski C Z, Chabik S and Rozenfeld B 1987 *Cryst. Res. Technol.* **22** K180
[22] Szymanski C Z, Chabik S, Pajak J and Rozenfeld B 1980 *Phys. Status Solidi* a **60** 375
[23] Puska M J and Nieminen R M 1983 *J. Phys. F: Met. Phys.* **13** 333
[24] West R N, Crisp V H C, DeBlonde G and Hogg B G 1973 *Phys. Lett.* A **45** 441
[25] Brandt W and Waung H F 1968 *Phys. Lett.* A **27** 700
[26] Kishimoto Y and Tanigawa S 1982 *Positron Annihilation* ed. P G Coleman, S C Sharma and L M Diana (Amsterdam: North Holland) p 404
[27] Würschum R, Bauer W, Maier K, Seeger A and Schaefer H-E 1989 *J. Phys.: Condens. Matter* **1** SA33–48
[28] Manninen M, Nieminen R, Hautojärvi P and Arponen J 1975 *Phys. Rev.* B **12** 4012
[29] MacKenzie I K, Craig T W and McKee B T A 1971 *Phys. Lett.* A **36** 227
[30] Eckert W and Schaefer H-E 1989 *Positron Annihilation* ed. L Dorikens-Vanpraet, M Dorikens and D Segers (Singapore: World Scientific) p 407
[31] Bauer W, Briggmann J, Carstanjen H-D, Decker W, Diehl J, Maier K, Major J, Schaefer H-E, Seeger A, Stoll H and Würschum R 1989 *Positron Annihilation* ed. L Dorikens-Vanpraet, M Dorikens and D Segers (Singapore: World Scientific) p 579
[32] Schaefer H-E, Gugelmeier R, Schmolz M and Seeger A 1987 *Mater. Sci. Forum* **15–18** 111
[33] Frenken J W M and van der Veen J F 1985 *Phys. Rev. Lett.* **54** 134
[34] Larsson K-E 1983 *Phys. Chem. Liq.* **12** 273
[35] Nachtrieb N H 1976 *Ber. Bunsenges. Phys. Chem.* **80** 678
[36] Shiotani D 1982 *Positron Annihilation* ed. P G Coleman, S C Sharma and L M Diana (Amsterdam: North Holland) p 561

[37] Faber T E 1972 *An Introduction to the Theory of Liquid Metals* (London: Cambridge University Press)
[38] West R N 1979 *Positrons in Solids* ed. P Hautojärvi (Berlin: Springer-Verlag) p 89
[39] Gebhardt E, Becker M and Dorner S 1953 *Z. Metallk.* **44** 573
[40] Weiler W and Schaefer H-E 1985 *J. Phys. F: Met. Phys.* **15** 1651
[41] Feder F and Charbnau H P 1966 *Phys. Rev.* **149** 464
[42] Puska M J and Corbel C 1988 *Phys. Rev.* B **38** 9874