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Positron annihilation in Zr, Ti, and a $Zr_{0.53}Ti_{0.47}$ alloy

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The temperature dependence of the Doppler-broadened linewidth of annihilation photons from an integral ^{68}Ge positron source has been studied in Zr, Ti, and a $Zr_{0.53}Ti_{0.47}$ alloy in the temperature range $20 < T < 1500$°C. The results are discussed in terms of a normalized peak fraction L of the annihilation photopeak area. Both Ti and the alloy show a predominantly linear increase of L with T below their respective hcp$\rightarrow$bcc transformation temperatures T_θ. For $T > T_\theta$, a second linear, but more pronounced, dependence of L on T is observed, the onset of which, in Ti, is marked by a discontinuous increase in L at T_θ. A further feature, common to the Ti and Zr data, is an apparent tendency at $T < 400$°C for a third region with a reduced temperature dependence of L to develop with increasing high-temperature exposure. Aside from data associated with this latter region, the Zr results indicate a rather linear increase of L with T below 750°C. At higher temperatures, the response of L to T increases before showing evidence of a decline near the upper temperature limit of the experiment.

I. INTRODUCTION

In the past five years it has become apparent that thermally activated mass transport in hcp ($\alpha-$) Zr involves both interstitial and vacancy defects.[1-4] There is, however, a lack of clear information on either the energy of vacancy formation, E_v^f, or the energy of vacancy motion, E_v^m, in α-Zr.

Furthermore concerning the question of the vacancy properties of α-Zr is the long-standing puzzle of the defect structure of β-Zr. The latter is a member of that class of materials, typified by β-Zr, β-Ti, and γ-U, which are known as the anomalous bcc metals, see, for example, Refs. 5–7. The characteristic of abnormally facile mass transport in these materials has been discussed in terms of both extrinsic diffusion via dislocations[6,8] and intrinsic diffusion via vacancies.[7,9] The former mechanism may involve an abnormally high dislocation density, associated with the prior bcc phase transformation,[5,6] while the latter suggests an anomalously large intrinsic vacancy population.

In recent years it has been determined that positrons in metals tend to be trapped by defects. This feature forms the basis of E_v^f determinations in metals by positron annihilation measurements (PAM). See, for example, reviews by West[10] and Seeger.[11] In general PAM on well-behaved, or normal, metals tend to become sensitive to vacancies at $T_v \sim 0.54\,T_m$,[12] at which temperature, the vacancy concentration is $C_v \sim 3\times10^{-7}$. As the temperature is increased, the fraction of positrons annihilating at vacancies increases, until saturation may become apparent. This type of behavior results in an S-shaped temperature dependence of a suitable characteristic positron-annihilation parameter.[13]

The present studies of Zr and Ti were made with a view to probing the defect structures of both the hcp and bcc phases of these very similar materials. It was felt that the lower T_m of Ti, 1675 °C compared to 1852 °C for Zr,[14] would bring the full temperature range of the bcc phase closer to the operating range of the apparatus, ~20–1500 °C. The study of the $Zr_{0.53}Ti_{0.47}$ alloy was made with a view to extending results for the bcc phase to lower temperatures. The alloy, which exhibits the same crystal structures as the component elements, transforms to the bcc phase at 540 °C, more than 300 °C below the equivalent transformation for Zr or Ti and exhibits melting at 1650 °C.[15]

In this work the temperature dependence of the Doppler-broadened line shape of 511-keV annihilation photons from a ^{68}Ge positron source, has been measured in terms of a normalized peak fraction, L, of the annihilation photopeak area. The choice of L was made to denote the emphasis being placed on the low momentum component of the annihilation spectrum. (In the past[16-18] the letter S has been used, but not consistently,[16,17] to identify a parameter similar to L. It is felt that the use of S could lead to confusion with the usual entropy symbol.)

In essence, the present work shows essentially linear regions of temperature dependence of L in Ti and $Zr_{0.53}Ti_{0.47}$, with no clear indication of any significant effects from positron interactions with equilibrium vacancies. In Zr, contributions from the latter source, although not well-defined, are suggested by a tendency towards an S-shaped L vs T curve. Annealing effects, observed in bcc Ti, may be indicative of positron interactions with a metastable defect system formed at the $\alpha \to \beta$ phase transformation.

II. EXPERIMENTAL

A brief summary of the experimental procedure is outlined below; a more complete presentation of this section of work will be presented elsewhere.[19]

The Zr and Ti samples were supplied as "superpure grade" by Aremco, New York, and contained 0.52- and 0.29-at.% oxygen, respectively. The total metallic impurity levels were 0.04 and 0.01 at.%, respectively, for Zr and Ti. The oxygen impurity content of the alloy was 0.69 at.%. (The oxygen impurity levels were determined by the analytical section of the Chalk River Nuclear Laboratories General Chemistry Branch.)

The sample for the measurements consisted of two discs, with a foil of the corresponding material, containing ^{68}Ge, sandwiched between them; the whole assembly, 12 mm diam × 6 mm thick, was diffusion bonded under compression, prior to any measurements.

During the measurements, the specimens were maintained under a dynamic vacuum, with a pressure $\leq 5\times10^{-7}$ Torr. All measurements for a given material were done on the same specimen with a constant geometry with respect to the sample, its immediate surroundings, and the detector.

The Ge(Li) detector provided a resolution of 1.45 keV at the 478-keV ^{7}Be line under normal operating conditions. Typically, 10^6 annihilation photopeak counts were accumulated in a customary counting interval of 10^3 sec.

The energy window, ΔE, of the centrally located peak used to define L was fixed for each material (except for the second series of Ti measurements—see Sec. III C and Fig 1). It was chosen to optimize a measure of the change between the 511-keV spectra corresponding to the low- and high-temperature states of the material being studied; conventionally, these states tend to be associated with annihilation of positrons in the free- and defect- (vacancy) trapped states, respectively. [To assist a comparison with the present work, ΔE and the corresponding full width at half maximum (FWHM) of the ^{7}Be 478-keV line, as measured during the experimental runs, are shown in Table I along with the energy calibration of the analyzer and representative absolute values of L.]

The enhancement of the peak intensity by defect trapping is a reflection of the lower-electron momentum distribution experienced by positrons annihilating at the defects. In principle, application of the Doppler technique to defect studies may be considered analogous to the similar application of angular correlation measurements of positron annihilation photons at zero angle, see, e.g., Ref. 20.

TABLE I. Energy windows ΔE, energy calibrations K, ^{7}Be FWHM, and absolute L values (at 600 °C) corresponding to the experimental data of the present work.

Material	K (eV/channel)	ΔE (keV)	^{7}Be FWHM (keV)	L (at 600 °C)
Ti	81.8	0.90	1.49	0.2768 [a]
	80.2	2.00	2.38	0.4966 [b]
Zr	83.6	0.92	1.41	0.2889 [c]
$Zr_{0.53}Ti_{0.47}$	83.1	1.08	1.41	0.3357 [d]

[a] Average value for data points 1 and 3 (Fig. 1).
[b] Average value for data 4 and 5.
[c] Average value for data 1, 2, and 4 (Fig. 3).
[d] Average value for data 1 (Fig. 2).

III. RESULTS

A. General résumé

Because the results of the present work reveal some dependence on thermal history, it is felt that a fairly detailed account of the chronology of the measurements might prove useful, both with regard to their discussion and with respect to any future comparisons with this investigation. This will be prefaced by a brief, general summary of the results.

The results of the present work are shown in Figs. 1–3. Both Ti and the Zr-Ti alloy show two distinct regions of near-linear dependence of L on T, with the transitions from the low- to the high-temperature slopes occurring at the respective phase transformation temperatures, T_θ. The results for Ti are further marked by a discontinuity in L at the phase transformation and, in common with the Zr data, a tendency towards attenuation in the response of L to T at temperatures below 400 °C.

The results for Zr (Fig. 3) show broad agreement with the Ti data (Fig. 1); there are, however, differences of detail which cannot be confirmed with certainty by the present work. For example, data 2 and 3 (Fig. 3), while indicating a generally smooth temperature profile, does not exclude the possibility of a discontinuity at T_θ, as suggested by data 1 and vice versa. Also it is not clear whether the L, T curve for Zr is exhibiting an enhanced temperature dependence just prior to T_θ. In an attempt to resolve these questions, further experiments on Zr, including work on single crystals, are planned.[21]

It is noted (Fig. 1) that whereas the nominal value of T_θ for Ti is 882 °C,[15] the value of T_θ, as

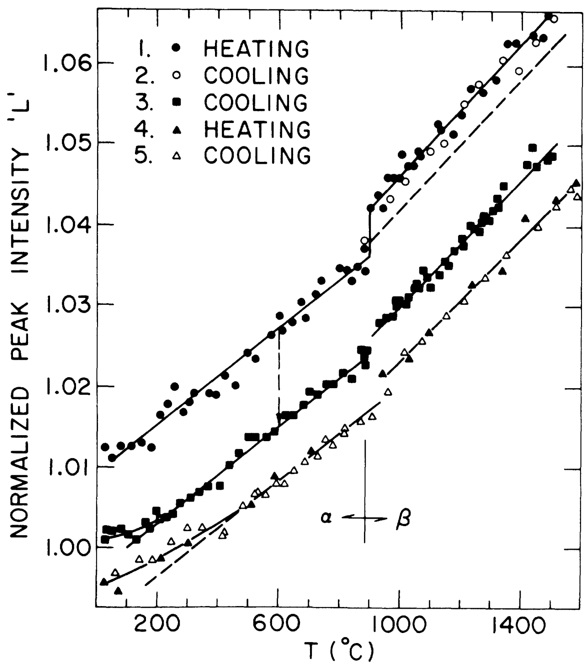

FIG. 1. Temperature dependence of the line-shape parameter L, describing the Doppler-broadened linewidth of annihilation photons from a ^{68}Ge source in Ti. Numbering of the curves reflects the chronological order of the measurements. Data labeled "heating" or "cooling" were taken in unbroken sequence. Within experimental error, data 3 coincide with data 1 from 20–880 °C. For ease of viewing, data 3 have been displaced downwards from data 1 by 1.2 scale divisions. Data 4 and 5 correspond to the given L scale expanded by a factor of 1.4, to make them comparable with data 1–3 (see Sec. III of the text).

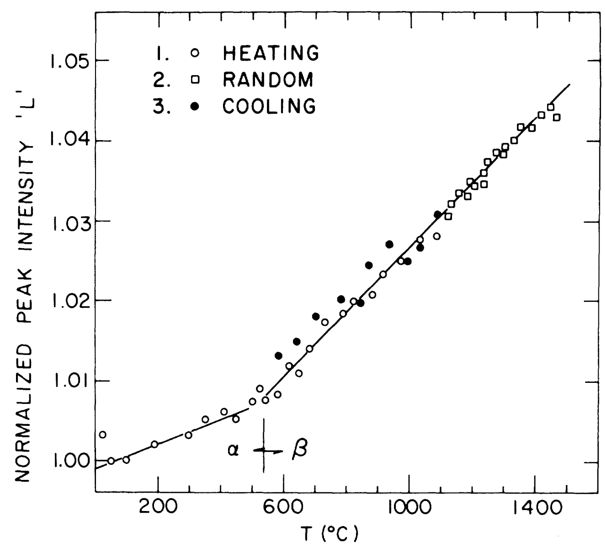

FIG. 2. Temperature dependence of the line-shape parameter L, describing the Doppler-broadened linewidth of annihilation photons from a ^{68}Ge source in $Zr_{0.53}$-$Ti_{0.47}$. Data labeled "random" were not accumulated in any regular sequence—otherwise the numbering and labeling in the legend have a meaning as in the Fig. 1 caption.

associated with the discontinuity in L, is (892 ± 8) °C and possibly even higher with increasing high-temperature exposure. This discrepancy may be attributable, at least in part, to stabilization of the α phase by dissolved impurities, e.g., oxygen[15] and carbon.[22]

B. Thermal treatment summary

The first experiments of the present work were done on Zr and occupied an interval of 22 weeks. The initial, unpublished measurements were made over a period of 3 weeks, following which an interval of 15 weeks lapsed, prior to the collection of the data shown in Fig. 3. The initial results were essentially as shown in Fig. 3, but with a much greater scatter; the 15-week lapse was largely devoted to equipment improvement. The next sample sutdied was $Zr_{0.53}Ti_{0.47}$ (see Fig. 2); here, as for the Zr data of Fig. 3, a period of four weeks was required for acquisition of the data. The last and most detailed study was on Ti (Fig.

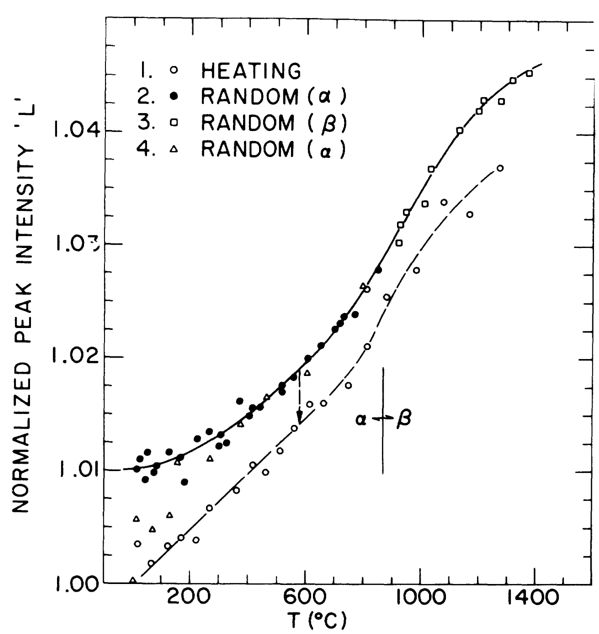

FIG. 3. Temperature dependence of the line-shape parameter L, describing the Doppler-broadened linewidth of annihilation photons from a ^{68}Ge source in Zr. Numbering and labeling have the sense described in the captions for Figs. 1 and 2. Data 1 coincide with data 2 and 4 in the vicinity of 600 °C, but they have been displaced downwards by 2.0 scale divisions for ease of viewing.

1) and involved three weeks of data accumulation. The largely accelerated data collection rate for the last work resulted from the initiation of automatic temperature control, which permitted almost continuous equipment operation.

C. Ti (see Fig. 1)

Following a 50-h anneal at ~880 °C, the sample was furnace cooled to 20 °C (8–10 h). The data points, 1, were then taken over a four day interval, following which, the sample was cooled to 900 °C over a period of 10 h while the experimental data 2 were taken. A three day anneal at 900 °C was succeeded by a one-step temperature increase to 1400 °C. Data 3 were then accumulated in the course of a four day cooling interval. After four days at 20 °C, data 4 were taken over a 14-h period. Next the sample was cooled to, and held at, 900 °C for 10 h prior to a one-step temperature increase to 1500 °C. Data points 5 were completed during a 24-h cooling to 20 °C.

It is to be noted (Fig. 1) that the L values for measurements 1, 2, and 3 correspond to identical measuring conditions, such that the absolute α-phase L values are reproduced at $400 < T < 900$ °C. The data 4 and 5, however, correspond to a much lower operating resolution of the detection equipment (see Table I) and a relatively high count rate $\sim 2\times 10^3$ cps in the 511-keV photopeak (see Sec. II). Thus the absolute L data, 1, 2, and 3, are not directly comparable with 4 and 5. It is also noted, notwithstanding optimization of the L parameter (see Sec. II), that the overall change in L for data 4 and 5 was only 3.5%, compared with 5.0% for data 1 and 3—presumably because of the lower operating resolution. For comparison with 1, 2, and 3, therefore, the L scale for points 4 and 5 has been expanded by a factor of 1.4.

D. $Zr_{0.53}Ti_{0.47}$ (see Fig. 2)

Preceding the Fig. 2 measurements, the sample was heated for 48 h at 530 °C. Data were then accumulated over a period of five days, during which time the temperature was held below 540 °C. The remaining data were gathered over a 22 day interval in which the temperature was maintained above 600 °C.

E. Zr (see Fig. 3)

Prior to the measurements, 1, the sample had been maintained at 20 °C for 15 weeks following a furnace cool to 20 °C from a 50-h β-phase exposure in the range 1000–1400 °C. Data 1 were amassed in a period of ten days. Before collection of data 2, the sample was furnace cooled following a 1-h exposure at 1100 °C. The results 2, 3, and 4 were gained over an interval of 18 days, during which the temperature was cycled at random, first in the α-phase, 2, then in the β phase, 3, and finally again in the α phase, 4. No very obvious trend with annealing treatment could be detected. The three very low points in 4 at 23, 69, and 131 °C, occurred in sequence near the end of the second α-phase anneal and were preceded and followed by "normal" data points at 164 and 276 °C, respectively.

IV. DISCUSSION

An overall view of the present work suggests that there are three identifiable regions of temperature dependence of L; these will be considered in order of ascending temperature.

The first region, region I, is associated with the low-temperature data for Ti and Zr (Figs. 1 and 3) which show a weak temperature dependence in the range 20–400 °C. The apparent lack of reproducibility of L suggests the extrinsic nature of this region. On the basis of comparisons with Ti and Zr, it seems probable that all the α-phase results for the alloy are to be identified with region I—see later remarks.

The second region (region II) concerns the α-phase data from the upper limit of region I to near T_θ. Because of the reproducibility of the results for Ti and Zr in this range (Fig. 1: 1 and 3; Fig. 3: 1, 2, and 4), in an absolute sense, as well as in their temperature dependence, this region is associated with intrinsic, bulk, or lattice annihilation events.

The third region (region III) concerns all measurements in the β phase. Clearly, for Ti the irreproducibility of the data in the long term (see later comments), indicates that this region, like region I, can be associated with extrinsic behavior. Contrary to observations in Ti (Fig. 1), the L values for the alloy tend to increase slightly with prolonged exposure (16 days) to temperatures in excess of 900 °C (see Fig. 2). The development of instability problems (associated with the detection equipment) during the last few measurements, makes it difficult to assess just how much significance should attach to the higher-L values in the range 600–1000 °C. With the exclusion of these higher-L values, the consistency of the β-phase data suggests association of region III for the alloy with intrinsic behavior. The data for β-Zr are not sufficiently well-defined to permit a decision regarding the nature of region III.

The apparent growth of region I in Ti and Zr, with increasing exposure to high temperatures indicates that the region may be associated with positron interactions with impurity-related defects, while the relatively low region I temperature range suggests the probable influence of association or precipitation events. The concentration of these defects may be expected to increase as the overall impurity content of the specimens increases[22]; O and N levels will increase with time, particularly at high temperatures.

Possibly some tendency towards a smearing of the phase transformation in Ti (Fig. 1) and the observation that all the α-phase data for the alloy, which had the highest starting impurity content of the materials studied, may be associated with region I (see later comments), could be regarded as consistent with the above description of that region.[22]

The linearity and extent of region II (up to T_θ) for α-Ti show no indication of significant interactions between positrons and equilibrium vacancies. A similar conclusion obtains for β-Ti and the bcc phase of the alloy.

As indicated in Sec. I, significant trapping of positrons in metals by thermally-generated vacancies, results in an overall S-shaped temperature dependence curve for the measured positron annihilation parameter.[13] Such a curve has been clearly observed in this laboratory by PAM on Al, using the Doppler effect,[23] and is suggested by the data (Fig. 3) for Zr.

An absence of positron-vacancy interactions has been noted previously for the alkali metals.[24] There it was suggested that lattice relaxation about the vacancy might reduce the probability of positron trapping. Similar comments have been made for other metals where no positron interactions with equilibrium vacancies have been observed.[11]

Since pressure-effect measurements on self-diffusion in β-Ti imply a strongly relaxed vacancy configuration,[25] the failure to observe positron-vacancy interactions there may not be surprising. The present results for α-Ti and the bcc alloy may imply that a relaxed vacancy configuration also occurs in these solid phases.

A combination of the empirical relation, $T_v \sim 0.54\, T_m$,[12] with an estimated T_m for α-Ti based on thermodynamic considerations[26] suggests that positron-vacancy interactions in Ti should become evident at ~700 °C; similar considerations lead to a corresponding figure of ~740 °C for α-Zr.[26]

While the α-phase data for Ti leave little doubt as to the absence of vacancy effects, the data for α-Zr do not permit clear resolution of this feature. It is noted that PAM (utilizing lifetime and angular correlation methods) do show sensitivity, at 20 °C, to vacancy defects formed in α-Zr by low-energy-electron irradiation.[27]

Region II for Ti and, to a lesser extent, for Zr, exhibits a well-defined slope. The reduced rates of change of L with respect to T (α_L) are 30×10^{-6}, 25×10^{-6}, and $19\times10^{-6}\,°C^{-1}$, respectively, for hcp Ti, Zr, and the alloy. The coefficient, α_L, is very similar to the temperature coefficient of volume expansion (α_V) for Ti and Zr; their respective values[28,29] of α_V are 30×10^{-6} and 21×10^{-6}. The correspondence between α_L and α_V continues into the β phase for Ti, where these quantities are 37×10^{-6} and 36×10^{-6}, respectively.[28]

Although a value of α_V is not available for the alloy, the low α_L value for the hcp phase, compared to those for α-Ti and α-Zr, suggests that for $T < 550$ °C, the alloy data may be more related to region I than the intrinsic region II.

While the close agreement between α_L and α_V in the present work may be somewhat fortuitous—in that α_L depends, to some extent, on the choice of L and the operating resolution of the photon detection system—it does, nonetheless, suggest that L is a volume sensitive parameter. Observations of a very similar nature have been made in angular correlation studies of positron annihilation in a number of other metals. There, a close similarity between the temperature dependence of the count rate at zero angle and α_V was observed.[30] These features were discussed in terms of an increasing probability of positron annihilation with valence, relative to ion-core, electrons.

The $\alpha \rightarrow \beta$ transformation, which also denotes the common boundary of regions II and III, is heralded in Ti and the alloy by a seemingly discontinuous increase in α_L and, in Ti, by a discontinuous increase in L (Figs. 1 and 2). For Zr, it is not clear whether the passage through T_θ involves a smooth or abrupt change in L and/or α_L (Fig. 3).

The results for Ti (Fig 1) show that the magnitude of the discontinuity at T_θ, ΔL, is dependent on thermal history (from the β-phase side, at least); but, within a limited time scale, ΔL may appear to be independent of heat treatment in the bcc phase, see 1 and 2, and 4 and 5. Data 1 and 3 show that with relatively long exposure to elevated temperatures (see Sec. II) L decreases. There is, however, no indication in the β phase that α_L is changing with annealing treatment.

Although the discontinuity in L at T_θ originates with the phase transformation, the basic factor, or factors, underlying the change and the subsequent shifts in L values are not known. Two possibilities suggest themselves. One, is that the above phenomena are associated at least in part with a defect structure originated by the $\alpha \rightarrow \beta$

transformation, such that prolonged heat treatment tends to anneal out the structure, leading to a time-dependent decrease in L. The other, is that all the Ti β-phase data are effectively intrinsic to the material and that the time dependence of L simply manifests a change in composition owing to the gradual uptake of impurities.[22]

The answer to this dilemma probably requires further work, aimed, perhaps, at determining whether or not there is a tendency towards a common level of annealed-state, or intrinsic, L values for β-Ti, as well as examining the reproducibility of ΔL at the transformation temperature. While such information is not presently available a few observations, which may be relevant to the issue, can be made.

Thus, for example, the close correspondence between α_L and α_V for Ti and α-Zr suggests that L is a volume sensitive parameter and accordingly may be expected to show a qualitative correlation with the volume change accompanying the phase transformation. Experimentally, the situation is quite the reverse. The changes, ΔV, in the specific volume at the transformation from hcp to bcc structure for Ti and Zr are, respectively, -0.4 and -0.8%,[28,29] while ΔL at T_θ is clearly positive for Ti and $\geqslant 0$ for Zr (Figs. 1 and 3). Thus it may be asserted that any simple correspondence between ΔL and ΔV at T_θ is overridden by effects due to the change in lattice structure, and/or the influence of defects.

Two features which may be viewed as lending some support to a defect contribution to ΔL at T_θ in Ti are the absence of any chronological effects on L in region II in Ti (or Zr) and the constancy of the region III results for the alloy (Fig. 2, data 2) over a long exposure period at high temperatures (see Sec. II). In both cases it might have been expected that uptake of impurities would have led to some change in L, were the high-temperature annealing behavior for Ti due simply to an increasing impurity content.

Since positrons are not observed to be interacting with vacancies in either α- or β-Ti, and since it seems highly improbable that any excess of point defects introduced by the $\alpha \to \beta$ transformation would last longer than seconds, it may be deduced that any positron-defect contribution to ΔL at T_θ, for Ti, is a reflection of positron interactions with extended defects, e.g., dislocations or grain boundaries. The contention above would lend experimental support to the protagonists of the formation of a metastable defect configuration at the hcp $\to$ bcc transformation for Ti. Such a phenomenon has been proposed to account—at least in part—for abnormally rapid mass transport in some bcc metals.[5,6] As discussed, further experimental evidence is required before any of the present results for Ti can be convincingly identified with defects contributing to diffusion in these anomalous bcc metals.

Only for Zr of the three systems studied here is there any tendency towards the S-shaped temperature-dependence profile of L associated with vacancy trapping. The effect, however, is very weak and within the limitations of the present data not well established.[21]

V. CONCLUSIONS

The temperature dependence of a normalized peak fraction, L, of the Doppler-broadened linewidth of annihilation photons from an integral ^{68}Ge source has been measured in Zr, Ti, and a $Zr_{0.53}Ti_{0.47}$ alloy in the temperature range 20–1500 °C.

The data show predominantly linear regions of temperature dependence of L for Ti and the alloy, with no indication of effects ascribable to the generation of equilibrium vacancies. The observation that L values in β-Ti may be subject to thermal history suggests the influence of a metastable defect structure originating at the $\alpha \to \beta$ phase transformation. The data for Zr are less well characterized and admit the possibility of effects due to thermally created vacancies.

In somewhat more detail, the results may be described in terms of three general regions of behavior. Region I, which is associated with effects due to impurity-related defects, occurs at $T < 400$ °C for Ti and Zr and at $T < 550$ °C for the alloy. This extrinsic region exhibits a relatively weak temperature dependence of L.

Region II is an intrinsic region, in which the reduced temperature dependence of L, α_L, is closely similar to the appropriate temperature coefficient of volume expansion, α_V, for Ti and Zr. Region II extends from the upper temperature limit of region I to T_θ for Ti and to near T_θ for Zr. Region II is believed to be masked by the extent of region I for the alloy.

The third region, region III, concerns all the β-phase measurements from T_θ. Although the Ti data again show equality between α_L and α_V in this region, the thermal history dependence of L points to its extrinsic character. The alloy results in region III appear to reflect predominantly intrinsic behavior. The data for Zr exhibit rather gentle curvature in region III and may be under the influence of effects due to equilibrium vacancies.

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[21]High-precision measurements on purer Zr specimens (than used in this work) confirm region I/region II behavior. For $400 < T < 960$ °C, $\alpha_L = 26\times10^{-6}\ C^{-1}$; deviations from this linear behavior are $< 0.05\%$ of L. See G. M. Hood and R. J. Schultz, International Conference on Properties of Atomic Defects in Metals, Argonne National Laboratory, Argonne, Ill., October 1976 (unpublished).

[22]Post examination of the specimens showed that carbon contamination from graphite spacers separating the samples from their Ta sheaths, had occurred in the Ti sample—it is not believed that this had any radical effect on the general nature of the Ti results, though it may have effected some changes of detail. Further comment on this aspect will be presented elsewhere [G. M. Hood and R. J. Schultz (unpublished)]. Similar carbon contamination was not evident in either the Zr or the $Zr_{0.53}\ Ti_{0.47}$ samples.

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