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The wide lower temperature internal friction peak and its positron annihilation study

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Abstract. A wide lower temperature internal friction (mechanical loss) peak was observed during heating and cooling processes in dilute Al–Mg solid solutions deformed by cold-rolling. By carefully decomposing using a nonlinear fitting program, the peak has a fine structure and is actually made up of three sub-peaks. After considering the behaviour of the three sub-peaks during the annealing process, we propose the 'vacancy mechanism' on the basis of interaction between dislocations and point defects for the origin of the three peaks. The thought was further supported by the corresponding positron lifetime spectrum study.

1. Introduction

The interactions between dislocations and point defects are a very important aspect of dislocation kinetics, especially for the interpretation and improvement of mechanical properties in crystalline solids. Internal friction (IF) is the dissipation of energy that occurs in a solid when the stress and the strain are not in phase. It can keenly provide us with useful information about this kind of interaction. In the cold-worked (twisted, stretched or wire-drawn) Al–Mg and Al–Cu solid solutions, seven IF peaks associated with the interactions were observed and designated as P_{L2}, P_{L1}, P_0, P_1', P_1'', P_2 and P_3 in order of ascending temperature [1]. Recently a lot of progress in the theoretical and experimental study on the room temperature (RT) IF peaks (P_0, P_1', P_1'') has been achieved in our laboratory [2–5], but the lower temperature IF peaks are little studied.

The positron annihilation technique is an ideal tool to probe this interaction between dislocations and point defects because it has intrinsic sensitivity to vacancies and solute atoms [6] as well as dislocations [7, 8]. Combining these two methods would provide us with a thorough understanding of this kind of interaction. In this paper, we have studied the properties of the IF peak and the corresponding positron lifetime spectrum in the dilute Al–Mg solid solution specimens. After comparing the results obtained by IF measurement and obtained by the positron lifetime spectrum study during the annealing process, we can conclude a 'vacancy mechanism' for the origin of the lower temperature IF peak.

2. Experimental procedure

The dilute Al–Mg alloy wires with 3 mm diameter are prepared by melting the required amounts of high purity Al (99.999%) and Mg (99.999%), which was made in China, in an

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induction furnace in argon atmosphere. The dilute Al–Mg alloy wires and pure Al wires are subjected to cold-rolling to the final thickness of about 0.8 mm, then cut to 8×8 mm^2 squares for the positron measurements and 70×8 mm^2 squares for IF measurements. The sample for IF experiments was loaded into an automatic inverted torsion pendulum immediately after cold-rolling and then cooled to liquid nitrogen temperature to start the experiment. The method we employ is free decay and the modulus is proportional to the square of the measurement frequency. For the annealing effect the sample was annealed *in situ* for 1 h. The samples for positron measurements were annealed isochronally in vacuum ($\sim 1 \times 10^{-4}$ Torr) for 1 h and furnace cooled to RT.

For positron annihilation spectrum (PAS) measurements, two identical samples were sandwiched with a 0.29×10^6 Bq ^{22}Na positron source, evaporated onto Kapton foil (about 5 μm thickness). The lifetime measurements were carried out at RT using a fast–fast coincidence system with resolution time 235 ps (FWHM) in operating conditions. The spectra, containing at least 5×10^6 counts each, were analysed by the Resolution and Positronfit computer programs [9]. A source component ($\tau_s = 509$ ps) with very small intensity ($I_s = 7\%$) is subtracted from each spectrum. The decomposition of the lifetime spectra allowed extraction of two components for all the deformed Al samples [6–8, 10].

3. Results and analysis

3.1. The fine structure of the wide lower temperature IF peak

A very wide lower temperature IF peak was observed in the dilute Al–0.12 wt% Mg sample deformed by cold-rolling during the heating and cooling processes, as shown in figure 1 (the change rate is 1 °C min^{-1}). Its corresponding modulus variation was plotted out.

It is worth noting that this peak is much wider than a standard Debye-type peak, which means it is probably made up of several sub-peaks. By using a nonlinear computer

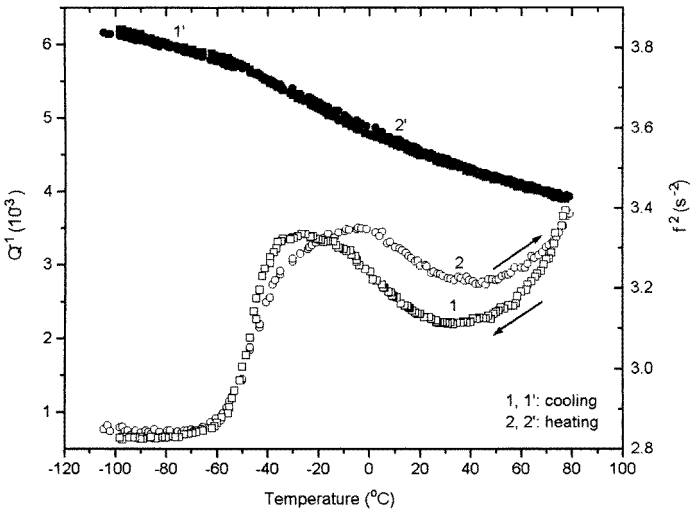

Figure 1. Appearance of the wide lower temperature internal friction peak in a dilute Al–0.12 wt% Mg solid solution sample deformed by cold-rolling during a heating and cooling process (rate is 1 °C min^{-1}). Hollow points stand for internal friction and solid points represent the corresponding square of measurement frequency (the same for all figures).

fitting program on the basis of Bevington's nonlinear fitting method [11], the IF curves are decomposed successfully into an exponential background and three sub-peaks with a lognormal distribution in relaxation time. The fitting results of figure 1 are shown in figure 4(a) (for the cooling process) and in figure 4(b) (for the heating process) and the results are so satisfactory that the curve passes through almost all the experimental data points in the range of experiment error (2×10^{-4}). The three sub-peaks are named peak X_1, X_2, and X_3 in the order of ascending temperature (the activation energies are kept constant as 0.8, 1.2 and 0.6 eV respectively). During the fitting, we keep the same origin and obtain a peak temperature shift for the every three peaks in the process of heating and cooling correspondingly. All the shifts contributed by the three peaks lead to a great peak temperature difference as shown in figure 1, which was theoretically discussed elsewhere.

Then we change to another Al–0.76 wt% Mg sample under lower oscillating frequency and higher heating and cooling rate ($2\,^{\circ}$C min^{-1}): this wide lower temperature IF peak was still observed, as shown in figure 2, but no such a lower temperature IF peak was observed in any high purity Al (99.999%) samples however the conditions were altered, as shown in figure 3. This suggests the solute Mg atoms play a very important role for the appearance of this peak. As we know, cold-rolling is a heavy deformation that produces a large amount of dislocations as well as supersaturated vacancies [12–14]. The lower temperature IF peak is therefore attributed to the interactions between dislocations and point defects resulting from the deformation because the Mg content is so far below its solid solubility in Al (about 10%) that the peak in terms of a precipitation process is excluded. But how the interaction works and what role the vacancies (or vacancy clusters) play can be answered by the further study, which is the aim of the remaining part of this paper.

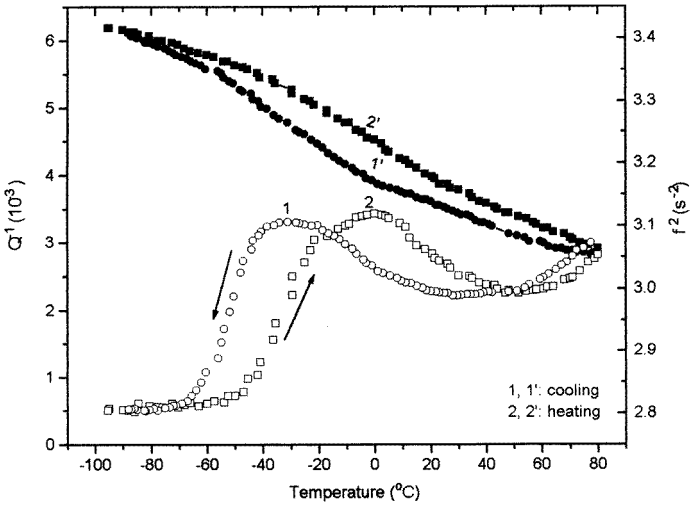

Figure 2. The wide lower temperature internal friction peak still appears under different oscillating frequency and different temperature change rate ($2\,^{\circ}$C min^{-1}) in an Al–Mg solid solution sample with different content (Al–0.76 wt% Mg).

3.2. Change tendency of the three sub-peaks during the annealing processes

We prepared another sample of dilute Al–0.12 wt% Mg alloy in the same way to study the three peaks' annealing effect. All the results were measured in the heating process,

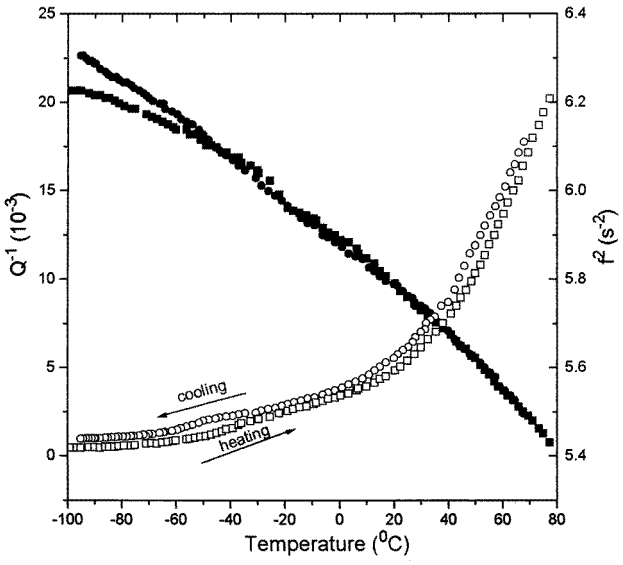

Figure 3. No such peak appeared in a high purity Al (99.999%) sample deformed by cold-rolling (the temperature changing rate is $2\,^{\circ}$C min^{-1}).

as shown in figure 5. There is a wide IF peak (the peak temperature is about $-2\,^{\circ}$C) for the 'fresh' deformation (immediately after cold-rolling) sample, as shown in figure 5(a). When the temperature is raised to $80\,^{\circ}$C, the sample is annealed *in situ* for 1 h and then cooled to $-110\,^{\circ}$C to start the heating measurement again. We found that peak height of the lower IF peak decreased and exhibits a wide 'peak shoulder' as shown in figure 5(b). In the same way, the sample is then annealed *in situ* for 1 h at different temperature: 100, 125, and $150\,^{\circ}$C successively, and the variations of IF are shown in figure 5(c), (d), and (e) accordingly. We can see that the peak height of the lower IF peak decreases with the increase of annealing temperature. When the sample is annealed at $200\,^{\circ}$C and above, the peak shoulder becomes smoothed and the lower temperature IF peak disappeared (which are not shown here). The changes of the three sub-peaks during the annealing process are also shown in the figure. In every fitting we keep the activation energy (1.2, 0.8 and 0.6 eV for X_1, X_2 and X_3 respectively) and distribution parameter β (a measure of the width of the lognormal distribution, i.e. the quantity of 2β is the width of the distribution at relative height $1/e$) constant. Distribution parameters of the peaks are set self-consistently to be 1.0, 5.2 and 7.1. According to Nowick and Berry [15], relaxation strength Δ is proportion to peak height Q_m^{-1} when β is constant. So we can quantitatively study the change of relaxation strength of three sub-peaks by comparing the peak height during the annealing process.

The variations of peak height and peak temperature for X_1, X_2 and X_3 are shown in figure 6. All the three peaks decrease after annealing at $80\,^{\circ}$C because the stresses resulting from cold-working are partially eliminated. But for X_1, the peak height decreases all the time with the annealing temperature; for X_2, its peak height changes little at 80–$125\,^{\circ}$C, and then decreases at higher temperature, and for X_3, the peak height changes a little during the annealing process. As we stated before, the cold-rolling is a heavy deformation that produces a large amount of dislocations as well as supersaturated vacancies and a fraction of these supersaturated vacancies is more likely to cluster to form divacancies [16]. They

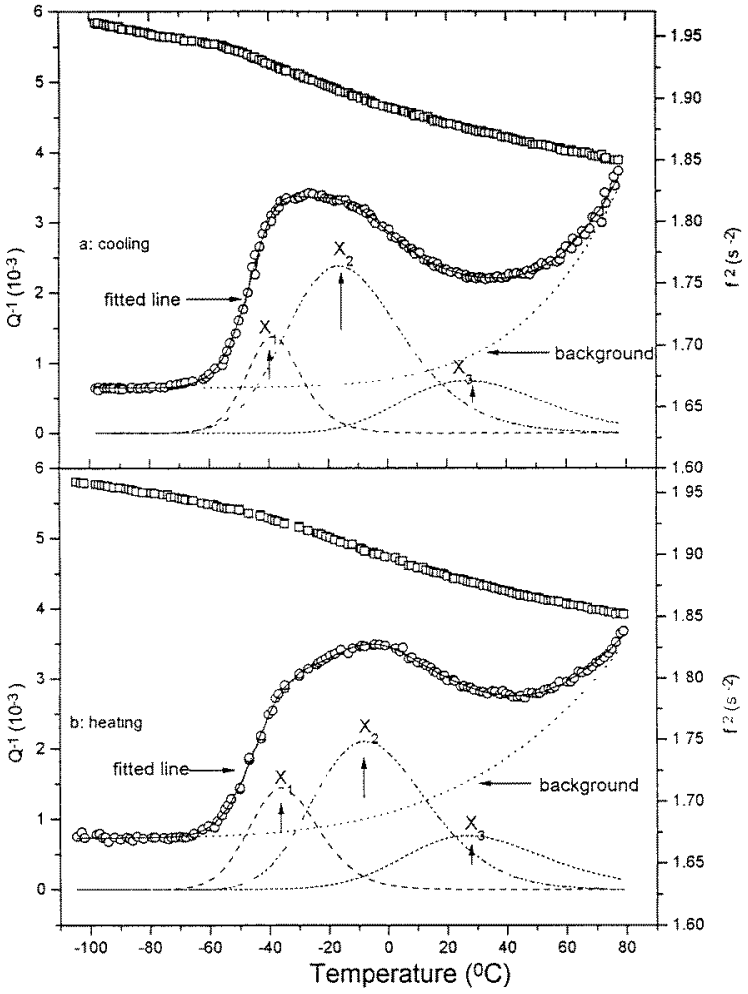

Figure 4. Fine structure of the wide lower temperature internal friction peak, which is made up of three sub-peaks X_1, X_2 and X_3. This is the results of figure 1.

are easily captured by Mg solute atoms to form 'Mg atom–divacancy' and 'Mg solute atom–vacancy' pairs [17, 18]. Based on the former results [1], X_1, X_2 and X_3 are proposed as the previously observed peaks P_{L2}, P_{L1} and P_1' , attributed to the interactions between dislocations and 'Mg solute atom–divacancy' pairs, 'Mg solute atom–vacancy' pairs and Mg solute atoms correspondingly. These complexes gather around dislocations to form something like a 'Cottrell atmosphere' and lag behind the movement of dislocations to give rise to internal friction. Under higher temperature, they would decompose because of thermal excitation, and the vacancies would diffuse to sinks to disappear. First, the 'Mg solute atom–divacancy' pairs begin to decompose into vacancies and 'solute Mg atom–vacancy' pairs, and then into Mg atoms and vacancies. So the peak height of P_{L2} decreases all the time while that of P_{L1} can receive this compensation to keep almost constant, and then decreases under high temperature. The results are in good agreement with the conclusions of Panseri *et al* [17], that solute Mg atoms in Al are able to trap vacancies at RT and at

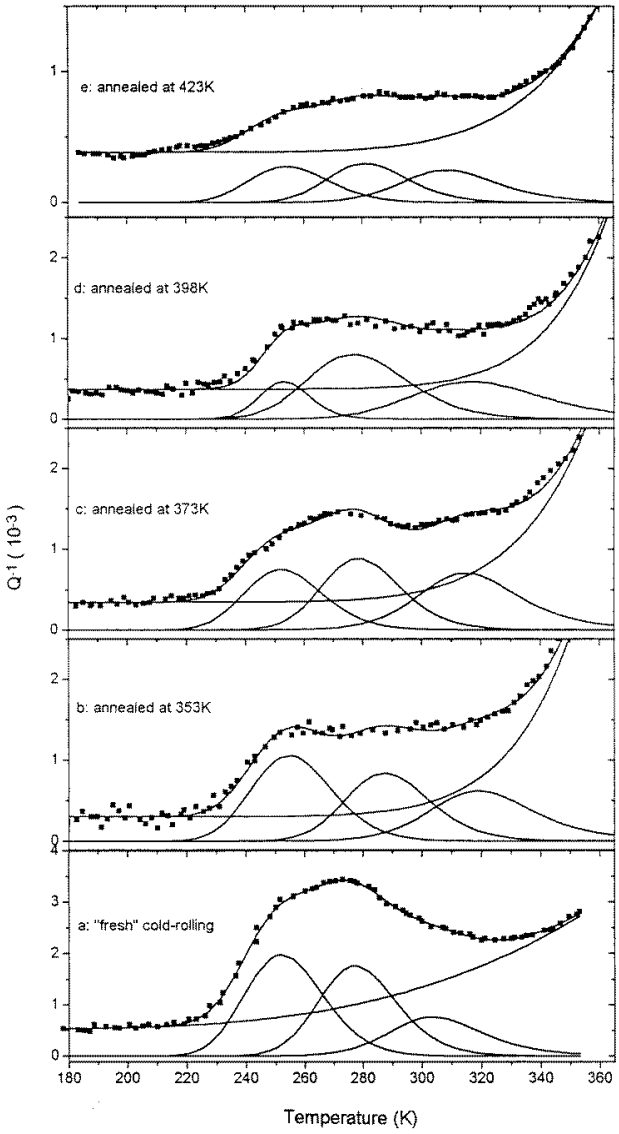

Figure 5. Annealing effect for the wide lower temperature internal friction peak and variations of the three sub-peaks during the annealing process. Points are measured and the lines are the fitting results.

about 120 °C vacancies can free themselves from traps. At the same time the fact that the diffusion activation energies for divacancies (0.48 eV) and for vacancies (0.66 eV) [18] are lower than the activation energies for P_{L2} (0.8 eV) and for P_{L1} (1.2 eV) suggests that they are associated with the diffusion of 'Mg solute atom–divacancy' pairs and 'Mg solute atom–vacancy' pairs as a whole. The peak height of P_1' changes little in the temperature range of 80–200 °C because it results from solute atoms diffusion along the dislocation core [2].

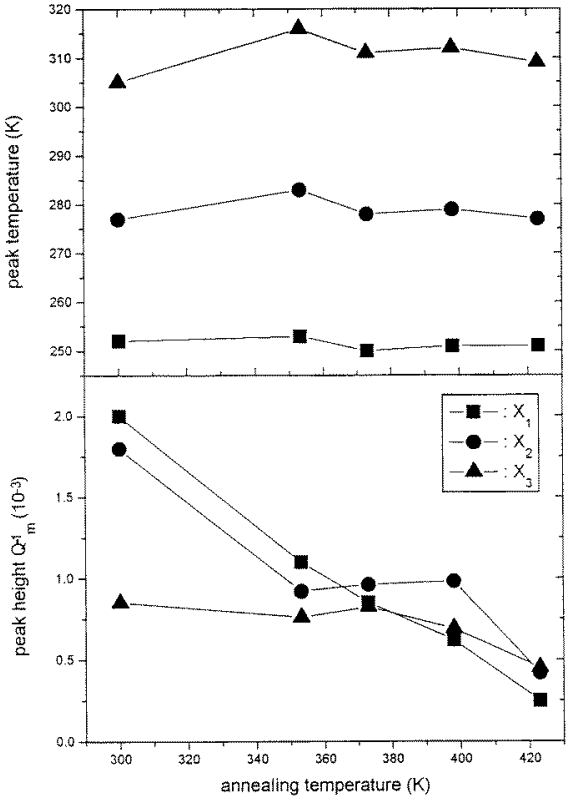

Figure 6. The quantitative variations of peak height and peak temperature for the three sub-peaks with annealing temperature.

3.3. Support from the positron lifetime spectrum study

To further verify our above 'vacancy mechanism' for the origin of the three peaks, the positron lifetime spectrum is measured with the specimen treated at the same condition as in the IF measurement. The results for the high purity Al sample and for the dilute Al–0.12 wt% Mg sample are shown in figure 7. According to the two-state trapping model, τ_1 and τ_2, are due to the annihilation of the free and defect state respectively, and the mean lifetime of the positron $\bar{\tau}$ is deduced from $\bar{\tau} = (\tau_1 I_1 + \tau_2 I_2)/(I_1 + I_2)$. In this paper we deal with what roles the defects play, so we discuss τ_2, I_2 and $\bar{\tau}$. Meanwhile τ_2, I_2 and $\bar{\tau}$ are the weighted averages of all sorts of defects (such as vacancies and their clusters resulting from short-range interaction of dislocations in the cold-rolling deformation [19]). For the Al sample, the bulk lifetime τ_f is about 163 ps, its monovacancy lifetime is about 238 ps [16] and the dislocation lifetime is about 230 ps [20]. Therefore the variation of its long-lifetime component τ_2 shows that the amounts of divacancies decrease with the increase of annealing temperature, and decompose wholly at about 150 °C. I_2 first decreases and then changes little at about 100–150 °C, resulting from the two opposite processes: on one hand the increase because of decomposition from divacancies, on the other hand the decrease because of the vacancy's migration to 'sinks' (such as dislocations, surface) to recombine. In this temperature range, meanwhile, the dislocations are also sinks for vacancies to vanish. They promote vacancies to sinks during the migration process, so

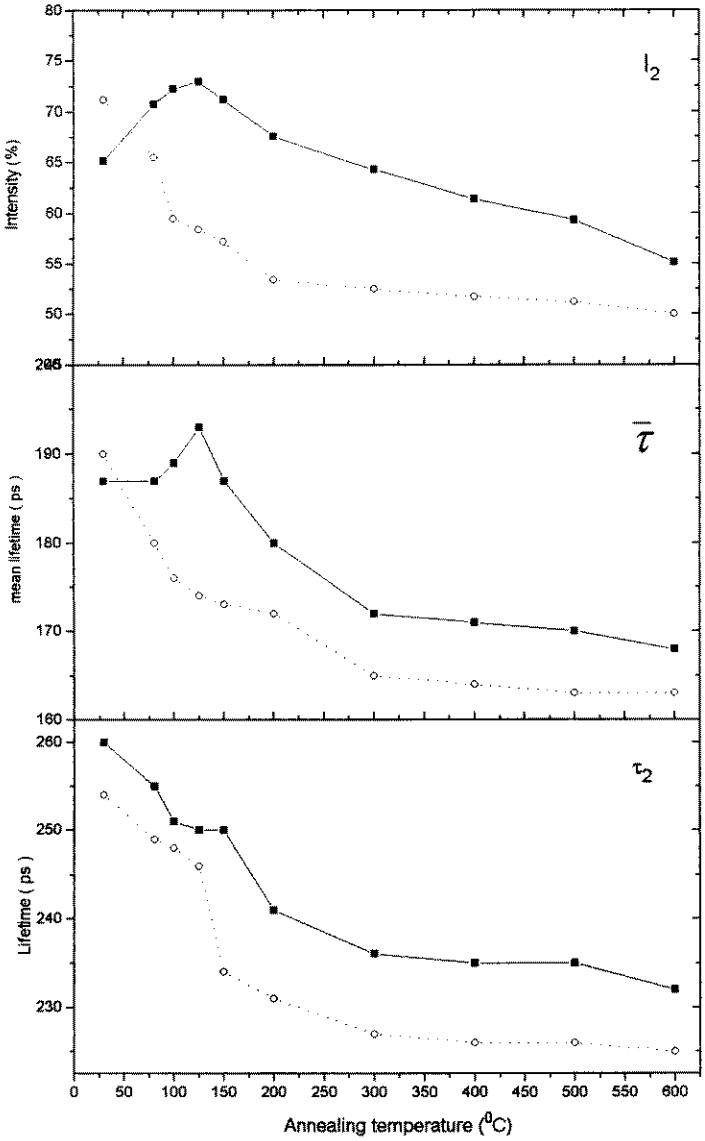

Figure 7. The change in the positron lifetime parameters on the dilute Al–0.12 wt% Mg solid solution sample and the high purity Al sample deformed by cold-rolling with annealing temperature. The samples are isochronally annealed for 1 h in a vacuum furnace at each temperature T. The solid points are for Al–Mg and the hollow points are for pure Al samples respectively. The lines (solid and dash) are drawn to guide the eyes.

the mean lifetime $\bar{\tau}$ decreases drastically (from 185 ps for 'fresh' deformation, to 180 ps for annealing at 80 °C, then to 176 ps for annealing at 100 °C). In the range of 100–200 °C the mean lifetime changes little due to these two contradictory processes. Under higher temperature 200–300 °C, the vacancies diffuse towards sinks and disappear gradually, so the $\bar{\tau}$ decreases quickly once more. When the annealing temperature reaches 300 °C and above ($\geqslant T_m/2$), the changes of lifetime due to point defects (such as super-saturated vacancies)

are almost entirely eliminated [21]. So the $\bar{\tau}$ is almost a constant 164 ps, which is in good agreement with the bulk lifetime of positrons in Al, and the drastic drop in τ_2 from the shoulder value to the saturated value corresponds to the lifetime of the positrons in dislocations.

But for the dilute Al–0.12 wt% Mg sample, there is a distinction from the Al sample, for vacancies and vacancy clusters are easily captured by Mg solute atoms to form a quite large number of 'vacancy–solute atom' complexes [17, 22, 23], which decrease the ability of a vacancy to trap a positron (but the positron lifetimes for these pairs are almost the same for the divacancies and for the monovacancy). That is, the formation of vacancy–solute atom complexes decreases the effective concentration of the vacancies 'seen' by the positron; at the same time the bulk lifetime of Mg is 230 ps (higher than that of Al) [16]. So τ_2 and $\bar{\tau}$ are larger than those of pure Al samples. In the range of 80–150 °C, the 'Mg solute atom–divacancy' pairs and the 'Mg solute atom–vacancy' pairs have been dissolved. Despite the decrease of τ_2, the increase of I_2 makes the mean lifetime $\bar{\tau}$ increase. When the 'solute Mg atom–divacancy' pairs decompose completely, the mean lifetime $\bar{\tau}$ reaches a maximum. In the range of 150–300 °C, the vacancies diffuse toward sinks (such as dislocations, grain boundaries and surface) swiftly and disappear, so $\bar{\tau}$ decreases clearly with the rising annealing temperature. At last $\bar{\tau}$ is almost a constant 170 ps ($>T_m/2$). The change tendencies for τ_1, τ_2 and I_2 are consistent with the results of figure 1 in [20]. Obviously, the difference between the Al–Mg sample and Al sample (such as a maximum existing in I_2 and $\bar{\tau}$ in Al–Mg but no maximum in Al) can be interpreted in terms of divacancies and vacancies being captured by Mg solute atoms and then decomposed under thermal excitation. So the 'vacancy mechanism' for the peaks P_{L2} , P_{L1} and P'_1, due to the interactions between dislocations and 'Mg solute atom–divacancy' pairs, 'Mg solute atom–vacancy' pairs and Mg solute atoms respectively, are confirmed in the experiment of the positron lifetime spectrum.

4. Conclusions

We have systematically studied the interactions between dislocations and point defects in the lower temperature range by means of the internal friction method and positron lifetime spectrum of a high purity Al specimen and dilute Al–Mg solid solution specimens with different Mg contents. The results can be summarized as follows:

(a) The solute atoms (such as Mg) and heavy deformation (for example, cold-rolling) to produce a large amount of dislocations and supersaturated vacancies (a fraction of them will cluster to divacancies) are two conditions for the appearance of the wide lower temperature IF peak, which is actually made up of three sub-peaks: P_{L2}, P_{L1} and P'_1.

(b) The supersaturated vacancies and divacancies are easily captured by solute atoms to form complexes, so P_{L2}, P_{L1} and P'_1 are tentatively associated with the interactions between dislocations and 'Mg solute atom–divacancy' pairs, 'Mg solute atom–vacancy' pairs and Mg solute atoms respectively. This 'vacancy mechanism' is well supported by the change tendency of the three peaks during the annealing process and further confirmed by the maximum appeared in the I_2 and $\bar{\tau}$ curves of the Al–Mg sample.

The theory aspect of the interactions between dislocations and these complexes as well as the determination of other parameters needs further study.

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