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by

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Low-temperature Effects in Metals as Observed by Positron Annihilation*

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Recently published experimental positron annihilation data in metals have demonstrated exceptional temperature-dependent behavior in the prevacancy region. However, the magnitude of these effects seems at present to scatter widely among different observers. New lifetime and Doppler-broadening results for Au between 30 and 590 K are presented and compared to results obtained in other laboratories. Doppler-broadening results obtained for Cu in the temperature region 300 to 1310 K demonstrate that some temperature-dependent structure observed in the prevacancy region is related to the thermal history of the sample.

In several recent observations of the temperature dependence of positron lifetime or lineshape parameters in presumably well-annealed metals it has been found that the temperature coefficient of the positron parameter decreases rapidly with decreasing temperature for temperatures in the prevacancy region (e.g., [1,2]). This behavior cannot be explained in a simple way from the theory of the positron Bloch-state, even when both lattice expansion and phonon interactions are taken into account. A generally acceptable explanation for the temperature dependence of the positron parameters in the prevacancy region has not yet been found.

Recently, Smedskjaer et al. [3] suggested that the temperature dependence of the lifetimes and lineshapes found in annealed Au for temperatures below 250 K by some observers could be due to positron trapping in extrinsic defects (e.g., impurities, dislocations). It was shown there that the temperature coefficients of the positron lifetime in Au measured by Herlach et al. [1], McKee and McMullen [4] and Smedskjaer et al. [3] were in good agreement for temperatures above 300 K, while discrepancies beyond those from statistical errors were found for temperatures below 300 K. In Fig. 1 a comparison is made among the lifetime data for Au of Smedskjaer et al. [3] (open circles), Herlach et al. [1] (shaded band) and McKee and McMullen [4] (single square point at $\sim$100 K). By subtraction of a constant from the data of [1] and [4], these lifetime measurements have been made to coincide at room temperature. Above 300 K all three data sets are close to the line shown and only the data from [3] are therefore presented. Below 300 K the differences among the data are apparent. In Fig. 2 a similar comparison between the Doppler-broadening data of Herlach et al. [1] and Smedskjaer et al. [3] has been made by scaling the data of [1] to the data of [3] for temperatures $\gtrsim$250 K (for details of the comparison, see [3]). It is again clear from Fig. 2 that a difference exists between these data for temperatures below 250 K. Both Fig. 1 and Fig. 2 suggest the presence of positron trapping in the sample of Herlach et al. [1] having yielded positron-annihilation parameters with significantly higher values than observed in [3] for temperatures in the prevacancy region, and one might suspect a similar but smaller effect in the sample of McKee and

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McMullen [4] (see Fig. 1). At present, the nature of the defect trap for the positron responsible for this behavior is undefined except that it must be extrinsic, since it does not appear to be inherent to Au independent of the state of the sample. Since even well-annealed metals contain both impurities and dislocations, and because the positron trapping appears at low temperatures, one must consider the possibility that weak positron traps associated with these extrinsic defects might have caused these effects.

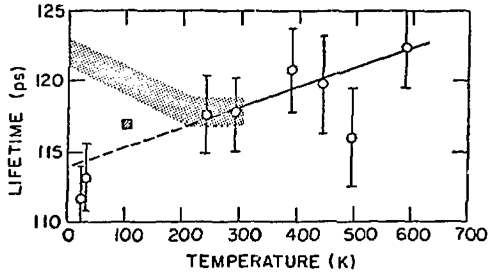

Fig. 1. Comparison of lifetime data [1,3,4] for Au (see text) [3].

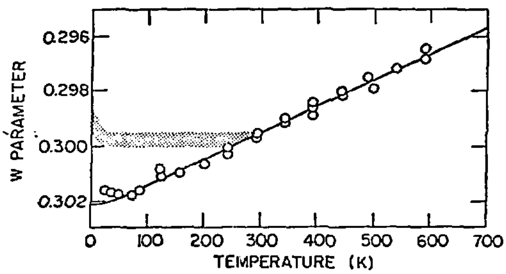

Fig. 2. Comparison of Doppler-broadening data [1,3] for Au (see text)[3].

The possible presence of positron trapping in the prevacancy region was further investigated in annealed Cu by Smedskjaer et al. [5]. During sample preparation procedures, in which the positron source-sample pakcage was diffusion bonded, a Cu sample containing a massless source [6,7] was heated to 420°C in a vacuum of 10^{-6} Torr while under a mechanical pressure of 5×10^6 N/m^2 for 12 h. The sample was then annealed at 950°C (0.9 of the melting temperature) for 3 h, before being carefully mounted in the vacuum ($\lesssim 10^{-6}$ Torr) chamber in which all subsequent annealing and positron annihilation measurements were carried out. The Doppler-broadening lineshape-parameter F was subsequently measured as a function of randomly selected temperatures below 890°C. The sample was then more fully annealed at 1020-1040°C for ∿2 h and F was remeasured for T $\leq$ 1040°C. The difference ΔF_B between these two F(T) measurements is shown in Fig. 3. The sample, subsequent to additional procedures described in [5], was then uniformly bent to produce a deformation of 3% and annealed at 850°C for ∿2 h. The lineshape-parameter F was again measured for T $\leq$ 850°C. Finally, the sample was annealed for ∿2 h at 1040°C and F was measured for T $\leq$ 1040°C. The difference ΔF_D between the latter two sets of F(T) measurements is shown in Fig. 4. In none of these cases was any observable recovery due to temperature cycling during the positron measurements detected. Therefore, neither Fig. 3 nor Fig. 4 represent annealing behavior, but simply represent the differences between essentially stable states of the samples involved.

The curves shown in Figs. 3 and 4 are differences in the least-squares fits of the trapping model to the respective data, taking both the extrinsic defects and the thermally generated vacancies into account [5]. From these fits it was found that the data in Fig. 3 correspond to a trapping of 5% of the positrons, while the data in Fig. 4 correspond to 2% trapping. In both cases it was determined that the lineshape parameter characteristic of annihilation in the extrinsic defects is less than or equal to that characteristic of annihilation in vacancies [5]. This seems to indicate that the electron density in the extrinsic defects is slightly higher than or equal to that in the vacancies.

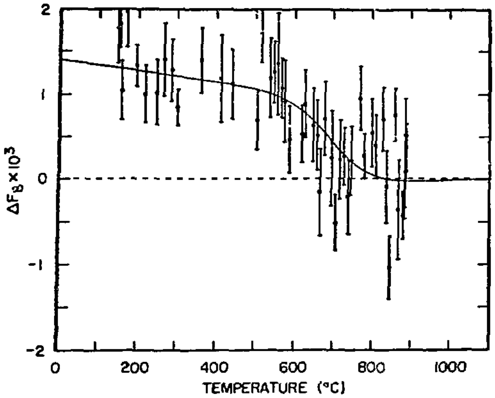

Fig. 3. ΔF_B vs. T for Cu (see text [5].

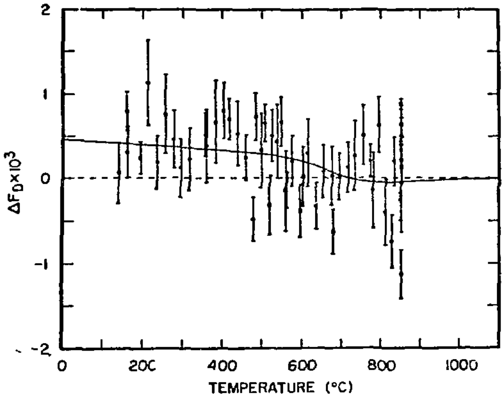

Fig. 4. ΔF_D vs. T for Cu (see text) [5].

In the case of Cu, as in the case of Au, it is not clear which specific extrinsic defect was responsible for the observed trapping, but either impurities or traps associated with dislocations could have contributed to these effects. However, in contrast to Au, the defects in these Cu samples were not weak positron traps, since the annihilation lineshape for these defects is similar to that of monovacancies and the trapping persisted to sufficiently high temperatures to be competitive with the onset of vacancy formation. If localization of positrons at impurity sites was responsible for the observed effects in Cu, then annealing at 1040°C could have removed the impurities from the bulk into the grain boundaries, whereas the 3% deformation could have led to a redistribution of the impurities, either through dislocation drag or by grain boundary motion during a recrystallization near room temperature. However, the more likely explanation of the results for Cu would appear to be positron trapping in dislocations or defects associated with dislocations. Annealing at 1040°C would then have effectively removed dislocation-associated traps through the decrease in energy of the dislocation network, possibly accompanied by a minor decrease in the dislocation density. This explanation might at first appear to be in conflict with those experimental observations [8,9] indicating that dislocation densities of 10^8-10^9 cm^{-2} are needed to cause observable positron trapping, while this Cu sample could be expected to have contained a dislocation density of 10^6-10^7 cm^{-2}. It should, however, be kept in mind that dislocations are a variety of defects, and that the dislocation networks present in the Cu sample need not have been identical to those produced in previous positron annihilation experiments (e.g., [10]) showing recovery for annealing temperatures of ∿400-500°C.

In conclusion, it has been demonstrated that significant positron trapping in extrinsic defects can be present in the prevacancy temperature region in well-annealed samples of Cu and Au. It would therefore seem prudent at present to question whether this is a more general phenomenon, which may already have been observed (e.g., [1,2,11]) in other metals as well.

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