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1989 J. Phys.: Condens. Matter 1 SA67

(http://iopscience.iop.org/0953-8984/1/SA/008)

J. Phys.: Condens. Matter **1** (1989) SA67–SA70. Printed in the UK

Gas densities in bubbles and positron annihilation characteristics

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Received 28 November 1988

Abstract. A survey of recent theoretical and experimental work on the relationship between positron annihilation parameters and densities of noble gas in bubbles is given. A combination of molecular dynamics simulations of the gas–metal interface and positron state calculations for a positron trapped at the interface has made possible a theoretical calculation of positron lifetimes in He bubbles in Al and Kr bubbles in Cu. For the Al–He system the theory is compared with experimental lifetime results for Al in which He bubbles are introduced by 600 MeV proton irradiation. For Cu–Kr, measurements were made on Cu containing ≃3 at.% Kr. In both cases good agreement between experiment and theory is found. In addition, one-dimensional angular correlation curves for He bubbles were determined from analysis of experimental results.

The positron annihilation technique (PAT) has recently emerged as a powerful technique for studying defects associated with He and other noble gases in metals and semiconductors (see, e.g., Jensen 1988). In order to interpret the PAT results and to extract quantitative information it is essential to establish the relationship between positron annihilation parameters and defect characteristics, especially for the bubbles filled with noble-gas atoms, which are readily formed due to the insolubility of noble gases in most materials. Hansen *et al* (1985) proposed a relation between lifetime and gas density based directly on the corresponding relation for bulk He fluids. However, recently this relationship has been studied theoretically in more detail (Jensen and Nieminen 1987a, b) and new experimental results on He bubbles in Al (Jensen *et al* 1988b) and Kr bubbles in Cu and Ni (Jensen *et al* 1988a) support the theory. In the present paper we provide a summary of this recent theoretical and experimental work on the dependence of positron lifetimes and angular correlation of annihilation radiation (ACAR) curves on gas densities inside bubbles.

The theoretical calculations (Jensen and Nieminen 1987b) are based on an extension of the corrugated mirror model of the positron surface state (Nieminen and Puska 1983) to a positron trapped at a metal–gas interface. The model was applied to a planar interface representing surface facets of a large bubble (i.e. with radii larger than 5–10 Å). The atomic geometry at the interface was determined in a series of molecular dynamics simulations. The results from these simulations in the form of density profiles

0953-8984/89/SA0067 + 04 $02.50

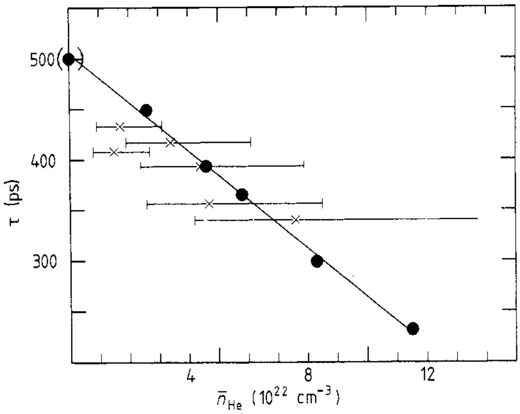

Figure 1. The lifetime for a positron trapped by a He bubble in Al versus the He density inside the bubble. Full circles are theoretical results (Jensen and Nieminen 1987a, b) and the line is a fit to these data. The crosses are based on experimental data for He bubbles in 600 MeV proton-irradiated Al (Jensen *et al* 1988b) with density estimates obtained from TEM by assuming all He in the samples to be in the visible bubbles.

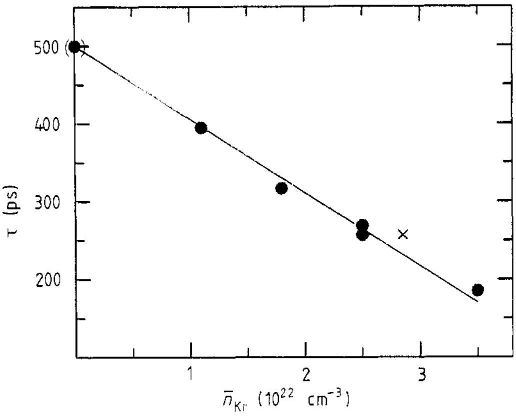

Figure 2. The positron lifetime as a function Kr density in Kr bubbles in Cu. Full circles denote theoretical results (Jensen and Nieminen 1987b). The line is a fit to these results. The cross is an experimental point for Cu containing ≃3 at.% Kr (Jensen *et al* 1988a) with the Kr density determined from electron diffraction.

perpendicular to the surface were then used to evaluate the influence of the gas on the positron state. One conclusion was that the positron binding energy for binding to the metal surface is increased by the presence of the gas at the surface. Thus the gas serves to stabilise the positron surface (interface) state. The calculations also show that the positron annihilation rate with metal electrons can be approximated accurately by the value for a bare metal surface ($\simeq 2\ \mathrm{ns}^{-1}$) while the annihilation rate with gas electrons depends on the gas density at the interface. This allows relationships to be obtained between the positron lifetime and gas densities; these are shown for the Al–He and Cu–Kr systems in figures 1 and 2. In both cases the theoretical results correspond to approximately linear relationships:

$$\text{Al–He:}\qquad \tau\ (\mathrm{ps}) = 500\text{–}23.5 n_{\mathrm{He}}\ (10^{28}\ \mathrm{m}^{-3}) \qquad (1a)$$

$$\text{Cu–Kr:}\qquad \tau\ (\mathrm{ps}) = 500\text{–}92.3 n_{\mathrm{Kr}}\ (10^{28}\ \mathrm{m}^{-3}) \qquad (1b)$$

where τ is the positron lifetime and n_{He} (n_{Kr}) is the average He (Kr) density at the interface which for a gas bubble corresponds to the density inside the bubble. Similar relationships are expected to hold also for other systems with different combinations of metals and gases (Jensen and Nieminen 1987b).

The theoretical results for the Al–He system are compared (figure 1) with experimental results (Jensen *et al* 1988b) for He bubbles in Al irradiated with 600 MeV protons which produce He via nuclear reactions. For as-irradiated samples the lifetime spectra showed saturation or near-saturation trapping into traps with lifetimes of 300–350 ps interpreted to be the He bubbles. Upon annealing the lifetime values increased reaching values of 400–450 ps for annealing temperatures around 800 K while the intensities of the He bubble lifetime component decreased. After completion of the positron measurements each sample was examined by transmission electron microscopy (TEM) in order to determine bubble sizes and concentrations. Independent estimates of the density of He inside the bubbles can then be obtained if all the He produced during

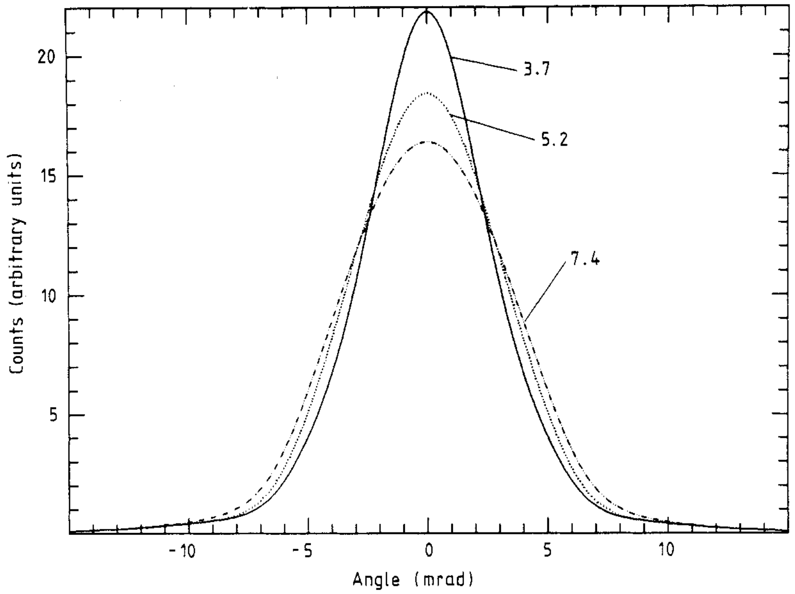

Figure 3. Angular correlation curves for positrons trapped in He bubbles in Al. The results are from an analysis of data for 600 MeV proton-irradiated Al (Jensen *et al* 1988b). The number indicated for each curve is the He density in units of $10^{28}\ \mathrm{m}^{-3}$.

irradiation is assumed to be in the visible bubbles. The density estimates for the experimental points in figure 1 have been obtained this way, while the positron lifetime values are the lifetimes of the He bubble component resolved from the lifetime spectra. The uncertainties in the density values were obtained from the estimated uncertainties on the TEM values for bubble radii and concentrations. The experimental points, despite the large uncertainties, are seen to reproduce the theoretically predicted trend.

Like the lifetimes, the angular correlation curves for positrons trapped in gas bubbles depend on the gas density. For He bubbles in Al this was established by analysis of ACAR curves measured on the same samples as used for the lifetimes experiments just mentioned (Jensen *et al* 1988b). The analysis of the parallel lifetime and ACAR results shows excellent consistency in terms of the fraction of trapped positrons determined from the two techniques and also in terms of a one-to-one correspondence between lifetimes and ACAR curve shapes for positrons trapped in He bubbles. This provides a relation between ACAR curve shape and He density, since the lifetime provides a measure of the He density via equation (1*a*). Figure 3 shows ACAR curves thus determined for three different He densities. It is seen that the ACAR curves become broader with increasing He density. No theoretical analysis of ACAR curves for gas bubbles is yet available.

The experimental result shown in figure 2 originates from a study of Cu containing about 3 at.% Kr (Jensen *et al* 1988a). The samples were prepared by a combined ion implantation and sputter-deposition method (Whitmell 1981). The as-prepared samples contain a high concentration of small bubbles which have been shown by electron diffraction to contain Kr in a solid phase at room temperature (Evans and Mazey 1985). The Kr lattice parameter obtained from the diffraction experiments gives a reliable determination of the average Kr density inside the bubbles. In figure 2 this density is correlated with the lifetime of the most long-lived component in lifetime spectra

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Table 1. Krypton densities (n_{Kr}) inside bubbles in Cu–Kr derived from positron lifetimes τ compared with densities estimated from TEM values for bubble radii r using the equilibrium bubble condition and the Kr equation of state. T is the temperature at which the samples have been annealed.

	PAT results		TEM results	
T (°C)	τ (ps)	n_{Kr} (10^{28} m^{-3})	r (nm)	n_{Kr} (10^{28} m^{-3})
As prepared	257	2.6	—	—
400	300	2.2	1.1	2.6
500	334	1.8	2.0	2.2
550	347	1.7	3.0	1.9
625	363	1.5	5.0	1.6
675†	374	1.4	10.0	1.2

† The TEM result was obtained at $T = 700$ °C.

correlated with the lifetime of the most long-lived component in lifetime spectra measured for the Cu–Kr samples. This result is seen to be in good agreement with the theoretical relationship between lifetime and Kr density. Isochronal annealing above 300 °C resulted in an increase in the positron lifetime showing a decrease in the Kr density. The Kr was then no longer solid at room temperature but in a fluid phase (Evans and Mazey 1985) and TEM results from similar Cu–Kr samples have shown that bubbles grow in size (Evans *et al* 1985). This is consistent with the fact that for equilibrium bubbles, i.e. bubbles where the internal gas pressure p is balanced by the surface tension γ, the bubble radius r is inversely proportional to the pressure:

$$p = 2\gamma/r. \tag{2}$$

Table 1 compares densities obtained from the positron lifetime results using equation (1*b*) with estimates based on the TEM results via equation (2) and the Kr equation of state relating pressure and density. The difference between the experimental conditions used for the two sets of measurements (different sample thicknesses and heating rates) precludes direct comparison but the two sets of densities are seen to be in good agreement.

The conclusion emerging from the results presented above is that quantitative estimates of gas densities inside bubbles can be made by PAT. Other methods have been used to address the gas density problem, but so far unambiguous density determination has been problematic (see, e.g., Donnelly 1985). PAT should therefore provide a major contribution to this area.

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