Professor H. Suhl for helpful conversations.

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[1]J. Ferraris, D. O. Cowan, V. Walatka, Jr., and J. H. Perlstein, J. Amer. Chem. Soc. 95, 948 (1973); A. N. Bloch, J. P. Ferraris, D. O. Cowan, and T. O. Poehler, to be published.

[2]L. B. Coleman, M. J. Cohen, D. J. Sandman, F. G. Yamagishi, A. F. Garito, and A. J. Heeger, Solid State Commun. 12, 1125 (1973).

[3]R. E. Peierls, *Quantum Theory of Solids* (Oxford Univ. Press, Oxford, England, 1953), p. 108.

[4]T. E. Phillips, T. J. Kristenmacher, J. P. Ferraris, and D. O. Cowan, Chem. Commun. 14, 471 (1973).

[5]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

[6]*The New York Times*, 25 March 1973, p. 43; Phys. Today 26, No. 5, 17 (1973).

[7]J. Bardeen, to be published.

[8]H. Fröhlich, Proc. Roy. Soc., London, Ser. A 223, 296 (1954).

[9]A. J. Epstein, S. Etemad, A. F. Garito, and A. J. Heeger, Phys. Rev. 25, 952 (1972); J. G. Vegter, J. Kommandeur, and P. A. Fedders, Phys. Rev. B 1, 2929 (1973).

[10]W. Kohn, Phys. Rev. Lett. 2, 393 (1959).

[11]For simplicity, we have adopted the commensurate case of one electron per molecule, where $q_0=\pi/a$. Our results will not be changed qualitatively for the noncommensurate case with q_0 close to π/a.

[12]J. Appel, Phys. Rev. Lett. 21, 1164 (1968).

[13]P. B. Allen, Solid State Commun. 12, 379 (1973).

[14]M. J. Rice and S. Strässler, to be published. We wish to thank Professor D. Scalapino for bringing it to our notice.

[15]W. Kohn and D. Sherrington, Rev. Mod. Phys. 42, 1 (1970).

[16]C. G. Kuper, Proc. Roy. Soc., London Ser. A 227, 214 (1955).

[17]B. R. Patton, Phys. Rev. Lett. 27, 1273 (1971).

[18]L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela 10, 1104 (1968) [Sov. Phys. Solid State 10, 875 (1968)].

[19]A. Griffin and V. Ambegaokar, in *Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964*, edited by J. G. Daunt *et al.* (Plenum, New York, 1965), p. 524.

[20]Preliminary measurements by B. T. Matthias, A. Lawson, and D. Johnston indicate such a big change in T_P with pressure.

[21]R. B. Aust, G. A. Samara, and H. G. Drickamer, J. Chem. Phys. 41, 2003 (1964).

Temperature Independence of Positron Trapping by Vacancies in Gold*

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Experimental data are presented which indicate that the trapping rate of positrons per unit vacancy concentration in gold is temperature independent.

The temperature dependence of positron trapping by vacancies in gold has been directly measured. The results show that below 0°C the positron trapping rate per unit vacancy concentration is independent of temperature. From the same data, but less directly, it appears likely that temperature independence continues up to at least 650°C.

The importance of this result arises from the increasing use of positron trapping to determine vacancy-formation energies E_v in metals.[1] The temperature dependence of trapping must be known if E_v is to be evaluated with an accuracy of better than about 10%.

The method was to fix the concentration of vacancies in gold, by quenching, and then to measure the positron lifetime at nitrogen and ice temperatures. The gold foil, of 99.99% purity from Johnson and Matthey, $0.4\times0.6\times0.010$ in.^{3}, was prepared by annealing for 4 h at 950°C in air and slowly cooling over about 12 h to room temperature. It was then etched. These pieces were clamped at each end in a light frame and quenched from 650°C by plunging them end first into water. The quenching rate was observed to be about 0.5×10^4°C per second for the first 125° while the foil was still in air and about 3×10^4°C per second thereafter in water. After quenching, these specimens were etched for a few minutes at room temperature and placed immediately in the apparatus which cycled them between ice temperature and 100°K. The few minutes at room temperature and the several hours at ice temperature and below during data collection should result in no significant annealing of monovacancies.[2] The change in positron lifetime between two fixed

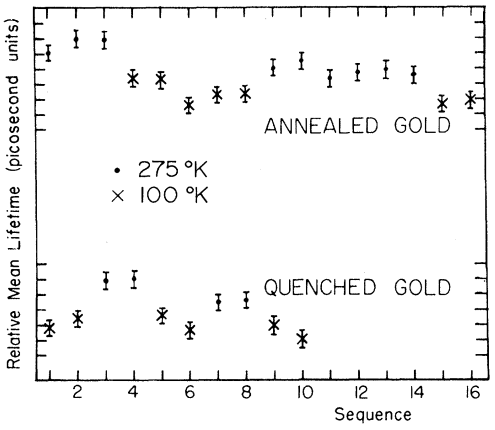

FIG. 1. Typical variation of positron lifetime in gold cycled between the two temperatures shown for annealed and quenched specimens. The centroid-shift technique used was designed to measure small changes in mean lifetime, not an absolute value. The scale of the ordinate is thus relative; picosecond units are marked. Statistical standard deviations are shown.

temperatures is shown in Fig. 1. Each point is the result of about 1 h counting time, and the apparatus required about an hour for a temperature change. The data were obtained by observing the shift in the centroid of the positron decay distribution as a function of temperature, a technique that yields quickly an accurate value of the change in mean positron lifetime. It can be seen that the mean life of positrons in the quenched gold at ice temperature is 2.3 ± 0.4 psec longer than at 100°K. For comparison, data from an annealed (12 h at 950°C) gold foil shows that the positron mean lifetime in gold at ice temperature is 2.2 $\pm$ 0.3 psec longer than at 100°K. The change in lifetime of positrons in annealed gold yields directly the effect of lattice expansion because the equilibrium concentration of vacancies is negligible at these temperatures. The gold specimen containing quenched vacancies shows essentially the same temperature dependence due to lattice expansion $[(2.3 \pm 0.4) - (2.2 \pm 0.3) = 0.1 \pm 0.5$ psec], implying directly no additional temperature dependence attributable to the trapping rate.[3] In what follows we describe the theory of the experiment and discuss the significance of the results.

The mean positron lifetime is $\bar{\tau} = \tau_v P_v + \tau_f (1 - P_v)$, where τ_v is the lifetime of positrons trapped in vacancies, τ_f the lifetime of free positrons, and P_v the fraction of positrons trapped and annihilating in the vacancies. The value of the mean lifetime corresponds to the observed centroid of the time spectrum[4] of positron decay. The positron trapping rate μC_v is easily shown to be given by $\mu C_v = P_v/\tau_f (1 - P_v)$, where C_v is the fractional concentration of vacancies.[5] The temperature dependence of μ, the trapping rate per unit vacancy concentration, can be obtained by differentiating the above equations to obtain

$$\frac{1}{\mu}\frac{d\mu}{dT} = \frac{1}{\tau_v - \tau_f}\frac{d\bar{\tau}}{dT}\frac{(\tau_v - \tau_f)^2}{(\tau_v - \bar{\tau})(\bar{\tau} - \tau_f)}. \quad (1)$$

The absolute values of τ_v, τ_f, and $\bar{\tau}$ had to be obtained from a slope analysis of the lifetime spectra of positrons decaying in the quenched and the annealed specimen. From these measurements we obtained $\tau_v - \tau_f = 60 \pm 10$ psec and $\bar{\tau} - \tau_f = 13 \pm 2$ psec. The value of the dimensionless last factor on the right-hand side of Eq. (1) is thus about 6 and is relatively insensitive to the value of $\bar{\tau}$. From the data of Fig. 1 we saw that the change in mean positron lifetime, apart from thermal expansion, was $\Delta\tau/\Delta T = 0.1 \pm 0.5$ psec/175°C. From additional data and systematic uncertainties, we suggest that the probable error in the above should be increased to about 1 psec. Thus we conclude that if μ varies as T^n over the temperature range involved, $n = 0.01 \pm 0.1$.

The trapping centers for the positrons are expected to be mostly monovacancies because of the low quench temperature. A 1-h anneal at 300°C of the quenched specimen removed all evidence of positron trapping, thus indicating a negligible contribution from quench-induced dislocations.

Information about the temperature dependence of positron trapping at higher temperatures can also be deduced from the data. The change in mean lifetime $\bar{\tau}$ in the specimen, measured when quenched and then when annealed, was 14 psec. This value should be compared with the results of Fig. 2 where we show the mean positron lifetime for equilibrium measurements in gold. It can be seen that between 0 and 650°C the mean lifetime changes by approximately 32 psec.

The difference between the 32 psec and the 14 psec is, in part, due to 8 psec from the lattice expansion discussed above. The remaining 10 psec can be attributed to either imperfect quenching, a temperature dependence of μ, or a combination of both. If we assume, rather unrealistically, a perfect quench from 650°C, the temperature dependence of μ would be described by $\mu \propto T^{0.7}$. However, in view of the slow cooling rate for the first 125°C, a perfect quench is certainly not realized and a vacancy concentration corre-

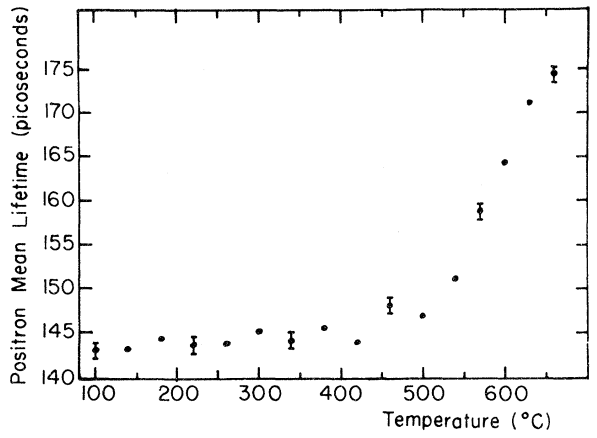

FIG. 2. Mean lifetime of positrons in gold (in thermal equilibrium) as a function of temperature. The centroid-shift method was used to measure the changes in mean lifetime with the accuracy indicated by the statistical standard deviations. A slope analysis of data at 0°C was used to fix the absolute scale of lifetimes with somewhat lower precision.

sponding to a considerably lower effective quench temperature would be expected. If μ were really temperature independent, Fig. 2 shows that the effective quench temperature would be 600°C. Thus it is reasonable to assume that most, if not all, of the 10 psec should be attributed to imperfect quenching and little, if any, to a temperature dependence of μ.

The experiment has thus shown that at low temperature, with a fixed concentration of vacancies, positron trapping is not a function of temperature. Assuming thermalization of positrons, this result shows that μ is independent of temperature, thus excluding both $T^{1/2}$ and $T^{-1/2}$ behavior which have been predicted.[6] The same temperature independence would be expected at higher temperature, and the experimental data support this conclusion.

Some of the variety of predicted behavior of $\mu(T)$, or equivalently μ(velocity), arises from different assumptions about the strength of the positron-vacancy interaction. A simple argument shows that a positron can interact with at least 100 vacancies before capture.[7] Thus the interaction may be considered weak. This, of course, is assumed in the calculation by Hodges,[8] which yields no velocity dependence based on his picture of a positron wave function spread out over a large volume encompassing many vacancy sites. A similar result may be obtained from a cross-section picture of a localized positron where the cross section far from resonance has an inverse-velocity dependence. Thus the product with positron flux is velocity independent. Such a cross section is usually interpreted as meaning that the probability for capture is proportional to the time the particle spends near the capture site.

It is interesting to speculate that if the second energy level[9] for the potential well is either a weakly bound state or a low-lying resonance in the continuum, then the velocity dependence might be very strong. However, an inspection of the known values of μ,[5] compared with the calculated potential well depth,[8] does not show much of a trend. Since the condition for a second bound state of a square-well potential of radius $a \sim 1.5$ Å and depth V, $2Vma^2/\hbar^2 > \pi^2$, implies a potential greater than about 15 eV, it is unlikely that trapping by monovacancies in metals should show a resonance capture process. Thus the temperature independence of positron trapping which we have measured for gold will probably apply to all metals.

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[1]See, e.g., the recent review by A. Seeger, J. Phys. F: Metal Phys. 3, 248 (1973).

[2]F. Cattaneo and E. Germagnoli, Phys. Rev. 124, 414 (1961).

[3]We expect lattice expansion to produce rather similar changes in the lifetime of both free and trapped positrons.

[4]The weak, longer-lived component customarily seen in positron lifetime spectra significantly affects determination of absolute lifetimes. It can, however, be ignored when determining centroid shifts as a function of temperature.

[5]B. T. A. McKee, W. Triftshäuser, and A. T. Stewart, Phys. Rev. Lett. 28, 358 (1972).

[6]D. C. Connors, V. H. C. Crisp, and R. N. West, Phys. Lett. 33A, 180 (1970); A. Seeger, Phys. Lett. 41A, 267 (1972).

[7]A positron of wavelength λ, velocity v, in free-travel time t, "sweeps out" a volume of order $\lambda^2 vt$. For appropriate values of λ, v, t, and vacancy concentration, this volume includes ~ 100 vacancies.

[8]C. H. Hodges, Phys. Rev. Lett. 25, 284 (1970).

[9]A resonance in a first bound state would not likely be observable, since the positron would be so loosely bound that its lifetime would not depart much from the free lifetime.