PHYSICAL REVIEW B VOLUME 10, NUMBER 12 15 DECEMBER 1974

Diffusion of Ag in Pb in the kilobar range*

A. Ascoli, L. Filoni, G. Poletti, and S. L. Rossi[†]
Centro Informazioni Studi Esperienze, Segrate, Milan, Italy

(Received 23 May 1974)

An accurate tracer-diffusion experiment was carried out on the system *Pb*(Ag) at pressures P between atmospheric pressure and 8.6 kbar. Activation-energy ΔH values between about 13.4 and 16.7 kcal/mole and an activation volume $\Delta V = 0.37$ atomic volumes were obtained. Although the general trend of the activation energy as a function of P extrapolates well the results published by Curtin *et al.* between 11.9 and 39.2 kbar, the ΔH-vs-P plot does not seem to be linear, and thus reinforces previous doubts on the singleness of the diffusion mechanism.

I. INTRODUCTION

The interpretation of pressure-diffusion results for noble-metal tracers in lead is in poor shape. Two groups of workers devoted themselves to such experiments. Ascoli and co-workers[1,2] specialized in fully hydrostatic piston experiments between 0 and 10 kbar; Vanfleet and co-workers[3-5] used a tetrahedral-anvil pressure apparatus up to 56 kbar, neglecting the above-mentioned pressure interval. Vanfleet and co-workers' results on silver and gold diffusing in lead appear to reject the purely interstitial and purely substitutional mechanisms, but they are not inconsistent with an interstitial-substitutional double mechanism.[4] In a subsequent paper[5] the same authors claim that a considerable fraction of Au and Ag impurities diffuse in Pb through the vacancy mechanism, whereas almost all Cu impurities diffuse in the same parent metal through the interstitial mechanism. Ascoli *et al.* found different dependences of diffusion parameters on pressure below and above a transition pressure, situated around 7 kbar for Au diffusing in Pb,[1] and between 1.75 and 3.38 kbar for Cu diffusing in Pb,[2] so giving evidence that the diffusion mechanisms are by no means simple, at least in the 0–10-kbar pressure range. In an attempt to generalize these conclusions, we decided to investigate the diffusion of Ag too in Pb in the same low-pressure range.

II. EXPERIMENT

The apparatus and method used for the Au-in-Pb (Ref. 1) and Cu-in-Pb (Ref. 2) pressure-diffusion experiments were adopted for the present work too, although pressure control was considerably improved.

A. Sample preparation

High-specific-activity $^{110}\mathrm{Ag}^m$ was chemically deposited from a silver-nitrate aqueous solution on one flat surface of well-annealed cylindrical 99.998%-pure Pb single crystals,[6] 1.1 cm in diam, about 1 cm long. The thickness of the Ag deposit turned out to be of the order of 1000 atomic layers or less.

B. Temperature range and pressure control

Each sample was annealed for a time interval ranging from 4 h, 10 min to 18 days, 19h, 40 min at a temperature between (153.9 ± 0.5) and (311.5 ± 0.5) °C and at a fully hydrostatic pressure between atmospheric and 8.6 ± 0.15 kbar. This narrow pressure fluctuation was obtained by equipping the primary oleodynamic circuit of the pressure amplifier with a hydraulic jack, powered by a pneumatic pump (fed in turn by the low-pressure compressed air supply of the laboratory) triggered by an on-off electric signal from a minimax manometer inserted in the primary oleodynamic circuit itself. The secondary pressure-transmitting fluid was glycerine. The usual controls (wax cones under the amplifier piston mushroom were undeformed after each experimental run, and the "unsupported-area" deformation of the low-carbon steel gaskets was roughly proportional to nominal applied pressure) ensured that, as in both the previous works,[1,2], pressure was effectively transmitted to the secondary circuit.

C. Temperature control and nominal diffusion temperature and time

Temperature was controlled by a Leeds & Northrup DAT or an Electromax stabilizer and recorded by a Speedomax model AZAR recorder.

Since the measuring thermocouple is at atmospheric pressure in our apparatus, no pressure correction of the thermocouple calibration was needed. Preliminary checks[1] ensured that the measured temperature differed from the actual sample temperature by less than a few °C during the heating and cooling transients and by less than 0.1 °C at any steady temperature.

For each annealing, the rate of heating was so regulated as to reconcile the needs of short rise time and minimum initial overshooting. Total heating times turned out to be about 2 h or less.

Maximum initial overshooting was about 10 °C, but in most cases it was confined within a few °C. The resulting temperature oscillation was in all cases strongly damped. At the end of the annealing, the pressure apparatus was lifted from the furnace and dipped into a forced-water-cooled oil bath. Total cooling times turned out to be $1\frac{1}{2}$ h or less; nominal diffusion temperature T_d was defined as the arithmetic average of the temperature T versus time t function, recorded between "initial" and "final" diffusion times. Of course, since the dependence of D on T is not linear, arithmetic averaging of T to obtain T_d is not rigorous, and is only justified by the small differences between averaged T values. The "final" time was always well defined on the T-vs-t records, since quick cooling in the oil bath produced a drastic change of dT/dt. Conversely, the definition of the "initial" time required some empirical criterion, since the above-mentioned compromise among contrasting requirements led to the final part of the "heating curve" being joined to the "steady temperature" profile by an envelope of exponentials. T_d and initial time are therefore strictly interdependent and were chosen, for each annealing run, by assuming as the initial time that for which $T \geqslant 0.90T_d$ and $dT/dt = 1.25\times10^{-3}T_d$ °C min^{-1} (the derivative dT/dt being averaged over 5 min). Due to the Arrhenius-type dependence of D on T and taking into account the expected activation energy ΔH values and the shape of T-vs-t records, it was easily checked that heating- and cooling-time contributions to diffusion were negligible.

D. Analysis of diffusion penetration

After annealing, 40 to 150 slices, 20–50 μm thick, depending on activity and the obtained penetration, were microtome-cut from each sample, and counted singly or grouped in couples. The 660-keV γ peak from ^{110}Ag^m in the slices was detected by a 3×3-in. or a 2×2-in. NaI(Tl) crystal scintillator counter and analyzed by means of a 400-channel pulse-height analyzer. Owing to long half-life of the tracer [253 days (Ref. 7)], no correction was needed for decay during counting.

Each microtome-cut slice was weighed to better than 0.1% in order to ensure proper evaluation of the penetration depth and specific activity.

III. RESULTS

The area under the 660-keV peak from each slice or couple of slices was normalized to the weight of the slice(s) to give their specific activity. This specific activity I was fed, as a function of the distance of the slices from the tracer deposition plane, to an IBM 1800 computer, programmed to give the least-squares adjustment of $\ln I$ versus x^2 on a straight line and the penetration plot.

So rich was the activity and such a deep penetration could be obtained that all the penetration plots look as neat as that in Fig. 1, so that only five penetration annealings were performed at each pressure value, to give in turn nine Arrhenius plots as neat as those presented in Fig. 2. Table I collects the results of this work. Dt represents the slope of the straight line, determined by the above-mentioned least-squares adjustment. D_0 and ΔH values are obtained from the zeroth- and first-order polynomial coefficients of the best fit of D versus $1/T$ in a straight line obtained for each P value on a Tektronix model 31 table computer. The correlation coefficients of the best fits range from 0.9983 to 0.99999. The errors stated in Table I for P are mean quadratic errors of P values measured during all experiments at the same nominal pressure. The errors stated for ΔH are larger than those obtained by the formula

$$\Delta(\Delta H) = \left[\left(\frac{\partial \Delta H}{\partial D}\,\Delta D\right)^2 + \left(\frac{\partial \Delta H}{\partial T}\,\Delta T\right)^2\right]^{1/2} ,$$

(where ΔD and ΔT are the mean quadratic errors on D and T at each pressure) since these turned out to be smaller than the accepted intrinsic uncertainty of the method.

IV. DISCUSSION

A. Evidence of fast-diffusion component in some samples

Some of the penetration profiles showed an apparently constant "tail" beyond the Gaussian dis-

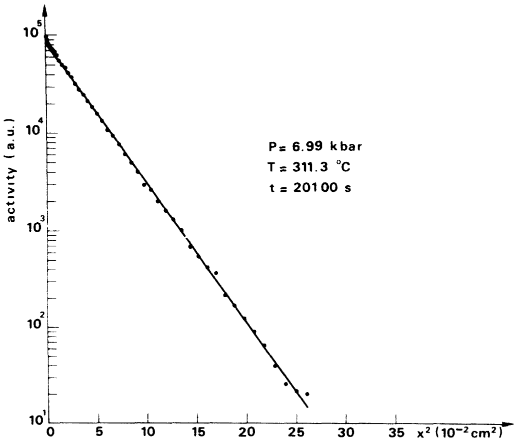

FIG. 1. Penetration profile at $T = 311.3$ °C and $P = 6.99$ kbar.

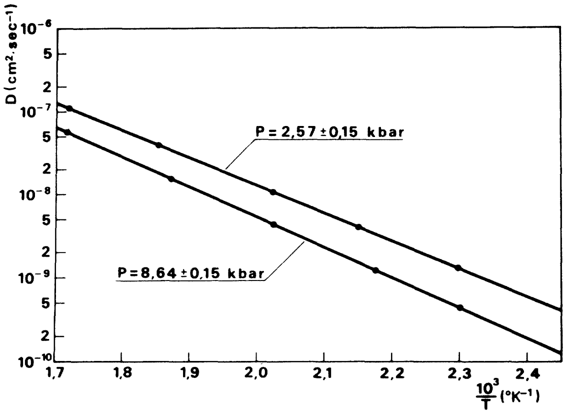

FIG. 2. Two typical Arrhenius plots.

tribution. The activity in the tail was never more than a few percent of that in the mainly Gaussian part; i.e., it was of the order of background. When it was not negligible, we subtracted it from the activity of all slices. A small fast-diffusion component has been found by several authors in various diffusing systems and is usually attributed to fast diffusion along dislocation pipes.[8,9]

B. ΔH vs P from separate Arrhenius plots, one at each P value

Figure 3 collects the ΔH values obtained in this work with those already published for Ag diffusing in Pb as a function of P. It can be seen that our ΔH value at atmospheric pressure checks well within error the value measured in air or *in vacuo* by Dyson *et al.*[10] The value measured by Seith and Keil[11] is higher, and is probably affected[12] by high Ag concentration (0.08%), and possibly by a less sensitive method.

It can be observed in Fig. 3 that, while the general trend of our ΔH-vs-P data extrapolates that of the data in Ref. 4 well, two of our points scatter from the curve well beyond experimental uncertainties. These two points, of course, give no acceptable evidence of a nonmonotonic trend of ΔH as a function of P. In fact, experiments at $P = 0.90$ kbar and $P = 2.57$ kbar were added to previously planned runs, to check the anomalous value of ΔH measured at $P = 1.63$ kbar, and their results do not confirm the anomaly. However, accurate reexamination of the laboratory records (quality of the samples, temperature, pressure, and specific-activity penetration records) and thorough consideration of all relevant experimental details supply no justification for errors larger than those shown in the figure. Where error bars are not shown, the uncertainties are smaller than the size of the points on the plot. Larger uncertainty at $P = 1.63$ kbar was estimated on the grounds of wider scatter of the points on the Arrhenius plot at that pressure than at other pressures, but we are left without an explanation for the two "anomalous" ΔH values.

Even leaving those two values aside, the line interpolating the ΔH-vs-P data is by no means a straight line and indicates a possible change in mechanisms with varying pressure (see also Sec. IVC). However the line presents no kink (as in Refs. 1 and 2) and therefore yields no definite evidence of two competitive mechanisms.

C. Best fit of all data on single equation

All (D, T, P) sets obtained in the present experiment were also best fitted to the equation of Weyland *et al.* [Eq. (6) of Ref. 4]. The results are as follows.

The difference in specific heats at constant pressure for diffusing Ag at the activated complex compared with its equilibrium position is $\Delta C_p = T_0(\partial \Delta S/\partial T)_p = 0.766R$, where R is the gas constant. The preexponential factor at $P = 0$ is $D_0 = 0.506\ \mathrm{cm^2\,sec^{-1}}$. The activation energy at $P = 0$ is $\Delta H = \Delta G + T_0 \Delta S = 15.290$ cal/mole, where ΔG is the Gibbs free energy, $\Delta S = \partial \Delta G/\partial T$ is the entropy of the process, and T_0 is the room temperature.

The activation volume (measured in terms of atomic volume V_0 of lead) at atmospheric pressure is

$$\frac{\Delta V}{V_0} = \frac{\partial \Delta G}{V_0 \partial P} = 0.366\,.$$

The temperature derivative of activation volume is

$$\frac{\partial \Delta V}{\partial T} = \frac{\partial^2 \Delta G}{\partial P\, \partial T} = -3.202 \times 10^{-3}\ \text{Å}^3\,{}^\circ\mathrm{K}^{-1}$$

$$= -1.062 \times 10^{-4} V_0\,{}^\circ\mathrm{K}^{-1}\,.$$

The pressure derivative of activation volume is

$$\frac{\partial \Delta V}{\partial P} = \frac{\partial^2 \Delta G}{\partial P^2} = 6.2711 \times 10^{-2}\ \text{Å}^3\,\mathrm{kbar}^{-1}$$

$$= 2.081 \times 10^{-3} V_0\,\mathrm{kbar}^{-1}\,.$$

Table II collects the data obtained by us and other workers.[3,4,10] Figure 4 represents the $\ln D$-vs-P curves obtained by imposing these best-fit results and three arbitrary constant values of T in the equation of Weyland *et al.*

Equation (6) of Ref. 4 is certainly based on firm theoretical grounds, and Table II shows that all values obtained for the analyzed parameters by fitting all our experimental data to that equation are of the correct order of magnitude (including D_0, considering that this parameter is extrapolated at a distance along the abscissa axis, which

TABLE I. Diffusion parameters obtained in this experiment.

P (kbar)	T_d (°C)	Dt (10^{-3} cm^2)	t (sec)	D (cm^2 sec^{-1})	ΔH (cal/mole)	D_0 (cm^2 sec^{-1})
0	298.4	4.2900	36 840	1.16×10^{-7}	14 520 ± 100	4.42×10^{-2}
	263.7	5.1273	93 780	5.47×10^{-8}		
	221.2	2.9432	183 600	1.60×10^{-8}		
	197.1	4.5184	596 400	7.58×10^{-9}		
	163.5	2.5094	1 104 000	2.27×10^{-9}		
0.57 ± 0.15	305.0	6.2042	46 500	1.33×10^{-7}	15 600 ± 100	1.15×10^{-1}
	270.4	5.9104	90 900	6.50×10^{-8}		
	238.2	4.1424	167 400	2.48×10^{-8}		
	208.0	3.4490	406 500	8.49×10^{-9}		
	182.9	2.8128	743 700	3.78×10^{-9}		
0.90 ± 0.15	303.8	4.3360	33 600	1.29×10^{-7}	15 010 ± 100	6.27×10^{-2}
	254.5	4.8644	129 900	3.75×10^{-8}		
	225.4	3.6155	258 900	1.40×10^{-8}		
	187.9	2.9754	594 000	5.01×10^{-9}		
	153.9	2.0517	1 656 000	1.24×10^{-9}		
1.63 ± 0.15	309.5	3.4348	31 800	1.08×10^{-7}	13 390 ± 500	1.23×10^{-2}
	276.9	3.1291	55 170	5.67×10^{-8}		
	245.0	3.2264	109 140	2.96×10^{-8}		
	224.7	2.2939	149 700	1.53×10^{-8}		
	199.5	1.5470	212 100	7.29×10^{-9}		
2.57 ± 0.15	308.3	2.0241	17 700	1.14×10^{-7}	15 250 ± 100	6.32×10^{-2}
	266.1	3.5309	88 500	3.99×10^{-8}		
	220.6	1.7060	157 200	1.09×10^{-8}		
	191.4	2.4531	602 400	4.07×10^{-9}		
	161.5	1.6438	1 269 600	1.30×10^{-9}		
3.48 ± 0.15	310.8	3.6155	33 840	1.07×10^{-7}	15 440 ± 100	6.67×10^{-2}
	279.4	2.8268	56 220	5.03×10^{-8}		
	249.3	2.3943	107 700	2.22×10^{-8}		
	219.9	1.5113	163 500	9.24×10^{-9}		
	202.9	1.2217	235 800	5.18×10^{-9}		
4.85 ± 0.15	311.5	1.2347	15 000	8.23×10^{-8}	15 190 ± 100	4.24×10^{-2}
	277.9	2.8385	72 540	3.91×10^{-8}		
	252.4	2.1823	99 300	2.20×10^{-8}		
	219.5	1.0293	144 960	7.10×10^{-9}		
	205.3	1.0280	220 800	4.66×10^{-9}		
6.99 ± 0.15	311.3	1.5046	20 100	7.48×10^{-8}	15 640 ± 100	4.91×10^{-2}
	278.8	1.5231	54 660	2.79×10^{-8}		
	250.8	1.2017	91 200	1.32×10^{-8}		
	199.2	0.7652	264 960	2.89×10^{-9}		
	160.4	0.7062	1 146 180	6.16×10^{-10}		
8.64 ± 0.15	309.6	1.7958	33 060	5.43×10^{-8}	16 510 ± 100	9.19×10^{-2}
	260.3	1.6764	104 700	1.60×10^{-8}		
	220.6	0.7761	177 300	4.38×10^{-9}		
	186.1	0.6775	553 800	1.22×10^{-9}		
	161.4	0.4443	1 047 600	4.24×10^{-10}		

is three times larger than the experimental interval).

However, we have two main reservations to make about the use of this equation.

First, to use the equation, values must be assigned to such parameters as γ (the Grüneisen parameter), β (the volume coefficient of thermal expansion), κ (the isothermal compressibility), $\partial\beta/\partial T$, $\partial\kappa/\partial T$, and $\partial\kappa/\partial P$. Most of these parameters are known with less accuracy than our experimental data. Besides, γ must be assumed to be independent of P and T, and this introduces a further questionable approximation.[13] These approximations may be responsible for the fact that the ΔH and D_0 values we obtain by fitting our data to the above-mentioned equation are only of the correct order of magnitude, whereas the values we obtain for the same quantities from the Arrhenius plot at $P = 0$ are in much better agreement with other published results. Another explanation may of course be that the investigated P interval is too narrow to give an accurate determination of pa-

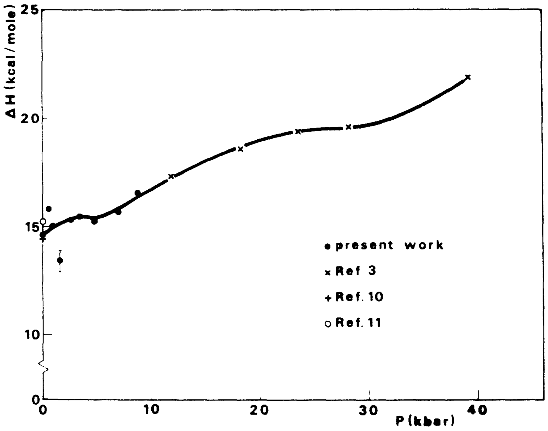

FIG. 3. Activation energy of the process, as a function of pressure.

rameters. However, we think that the main uses of this analysis are to check the general data consistency and to calculate the weighted averages of $\Delta V/V_0$, ΔC_p, $\partial\Delta V/\partial P$, and $\partial\Delta V/\partial T$ on the whole set of data.

The second reservation we have about this kind of analysis is of a more fundamental character. Weyland *et al.*[4] analyze their measurements assuming a single effective diffusion mechanism characterized by a possible pressure- and temperature-dependent activation energy and preexponential frequency factor. However, evidence of competing mechanisms, more complicated than interstitial and vacancy ones, keeps accumulating, even in the more open lattices which would be expected to favor simple interstitial mechanisms,[14] and finds general[9, 15-18] although controversial[19, 20] theoretical support.

The approach of assuming simple diffusion mechanisms even at the cost of drawing straight lines through other workers' experimental points (calling them scattered) was adopted in 1966 by Anthony *et al.*[21] Those authors compressed the ln$D$ scale of Fig. 3 of Ref. 1 until they obtained straight lines nearly parallel to the pressure axis, so hiding a nonlinearity which was evident in the curves. So doing, they disregarded the fact that a curvature, if borne out by experimental data, is best studied by choosing the x and y scales in such a way that the slope of the plot is nearly 45°. They even magnified the experimental uncertainties stated by the experimental workers, neglecting the fact that the authors of a work are undoubtedly in a better position than any reader to judge the probable errors of their individual data points. Weyland himself[22] has recently shown, on the grounds of experimental results by Kidson,[9] that the assumption of Ref. 21 is invalid for silver. Of course, this does not rule out a contribution of interstitial diffusion (which has recently[23] been extended to transition metals in Pb) among other mechanisms. What we reject is the assumption *a priori* that the mechanism must be single.

In fact, the activation volume we find, of the order of $0.4V_0$ (V_0 is atomic volume), compared with the theoretical estimate of the volume of motion of an interstitial in an fcc metal [$\Delta V_m = -0.01V_0$ (Ref. 24)] and of formation of a vacancy in Pb [$\Delta V_f = 0.55\ V_0$ (Ref. 25)] and with the results of high-pressure quenching[26] ($\Delta V_f = 0.53\ V_0$), annealing[27] ($\Delta V_m = 0.15\ V_0$), and self-diffusion[28] ($\Delta V_f + \Delta V_m = 0.64\ V_0 - 0.72\ V_0$) experiments, seems to rule out interpretations based exclusively on a pure interstitial or on a pure vacancy mechanism.

Furthermore, Weyland *et al.*[4] choose $\Delta C_p = 0$, on the assumption that the change in the number of degrees of freedom for the impurity at the activated complex compared to its equilibrium position

TABLE II. Measured parameters for diffusion of Ag in Pb.

Quantity and unit[a]	Present work Fit of all the data on equation of Weyland *et al.*	Arrhenius plot at $P=0$	Ref. 4	Ref. 10
D_0 (cm^2 sec^{-1})	0.506	0.045 ± 0.010	0.055 ± 0.013	0.046 ± 0.010
ΔH (cal/mole)	15 290	14 520 ± 100	14 630 ± 240	14 400 ± 500
$\Delta V/V_0$	0.366	...	0.34 ± 0.04	...
$\Delta C_p/R$	0.766	...	$[-0.5<(\Delta C_p/R)<0.5]$	...
$\frac{\partial\Delta V}{V_0\partial P}$ (10^{-3} kbar^{-1})	2.081	...	3.0 ± 0.8	...
$\frac{\partial\Delta V}{V_0\partial T}$ (10^{-4} °K^{-1})	−1.062	...	-1.9 ± 0.4	...

[a] See the text for definition of symbols.

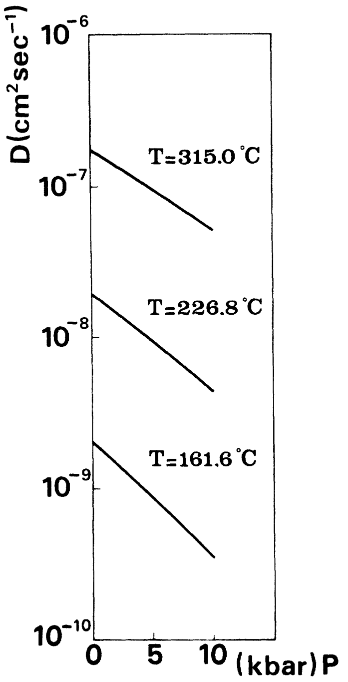

FIG. 4. ln*D* vs *P* dependence, at three arbitrary constant *T* values, obtained by the equation of Weyland *et al.* and our experimental results.

would not be expected to differ by more than one. This is certainly true (at the diffusion temperatures, which are well above the Debye temperature, and where therefore the equipartition law should hold) for a single mechanism and possibly for a few simple mechanisms acting competitively, but it is not necessarily true for a complex mechanism. In fact, the ΔC_p value resulting from the fit of our data is by no means equal to zero, thus contributing to the doubts about either simplicity or singleness of the mechanism(s) involved.

The curvature of our ln*D*-vs-*P* plots is, however, about

$$\frac{\partial^2 \ln D}{\partial P^2} \simeq -7.7\times 10^{-4}\ \text{kbar}^{-2} \quad \text{at } 315\,^\circ\text{C}$$

and

$$\frac{\partial^2 \ln D}{\partial P^2} \simeq -1.042\times 10^{-3}\ \text{kbar}^{-2} \quad \text{at } 162\,^\circ\text{C}\,.$$

It is, therefore, slight and negative, and it obeys the rule of Anthony *et al.*[21] Should a dissociative mechanism be responsible for the diffusion of Ag in Pb, the observed curvature would indicate that

$$0 < c_s/c_i < (D_s/D_i)^{1/2}\ ,$$

where c are concentrations and subscripts s and i refer to substitutional and interstitial, respectively. This would mean either that substitutional concentration is low in comparison with interstitial concentration, contrary to Kidson's conclusions for Ag in Pb, or that substitutional diffusion coefficients are higher than interstitial diffusion coefficients.

V. CONCLUSIONS

Fine pressure control ($\Delta P = \pm 0.15$ kbar), fully isotropic applied pressure (ruling out any unaxial component that could give rise to dislocation sweep), narrow steps between chosen pressure values, no temperature gradients along the sample (the whole pressure chamber is brought to temperature T), and small scatter of the experimental points on the penetration profiles (giving in turn small errors on measured D values) allow us to draw a certain number of conclusions from this experiment. We list first those following the discussion of Sec. IV C, then those following Sec. IV B.

First, a definite readily measurable negative curvature appears in the ln*D*-vs-*P* plot. This is consistent with a dissociative mechanism only if $c_s < c_i$ or $D_s > D_i$.

Second, the analysis of Weyland *et al.*, taking into account that a (the lattice parameter), ν (the average frequency of oscillation), and ΔG are functions of P and T, is certainly suitable for giving the best fit of large data sets, measured in well-diversified experimental conditions, provided that all relevant experimental conditions are taken into account properly. The fact that the coefficients of some of the terms in the equation are known with less accuracy than the diffusion data themselves and are not constant might prove of small importance, since they only influence correction terms. However, the fit of our diffusion parameters to other published data is worsened by the use of the unitary analysis of Weyland *et al.* (see Table II), and this might be explained by the possible influence on the results of sample characteristics and experimental method, which are usually disregarded in the interpretation of single Arrhenius plots, but may become relevant for a unitary analysis (e.g., dislocation density and distribution, single-crystal growth conditions, and cutting procedure).

Third, our value for the activation volume ($\Delta V = 0.366 V_0$) is intermediate between that for interstitial motion ($\Delta V_m = -0.01\ V_0$) and that for substitutional migration ($\Delta V_f + \Delta V_m \simeq 0.7 V_0$), and suggests a concurrence of mechanisms.

Fourth, even though neglecting the "scattered" points, the ΔH-vs-P plot is not linear. This is taken as a warning against drawing diffusion models from experimental results obtained at too widely spaced pressure values.

The fifth observation regards the two "scattered" points on the ΔH-vs-P plot. Although, of course, they offer no support to speculation, yet experimental accuracy and careful control of experimental conditions confirm that we lack a suitable explanation for their behavior. This may also suggest caution in adopting simple diffusion models, especially in view of improved parameter control.

Our over-all conclusion is that a firm proof is missing for both singleness and plurality of mechanisms for the diffusion of Ag in Pb. Since simple models are generally easier to demonstrate than complex models, this uncertainty is likely to hide complexity.

---

*Work coordinated with the Gruppo Nazionale Struttura della Materia program of the Italian National Research Council.

†Present address: Silverstar Ltd. S.p.A., Milano, Italy.

[1] A. Ascoli, B. Bollani, G. Guarini, and D. Kustudic, Phys. Rev. 141, 732 (1965).

[2] A. Ascoli and G. Poletti, Phys. Rev. B 6, 3681 (1972).

[3] H. R. Curtin, D. L. Decker, and H. B. Vanfleet, Phys. Rev. 139, A1552 (1965).

[4] J. A. Weyland, D. L. Decker, and H. B. Vanfleet, Phys. Rev. B 4, 4225 (1971).

[5] C. T. Candland, D. L. Decker, and H. B. Vanfleet, Phys. Rev. B 5, 2085 (1972).

[6] A. Ascoli and E. Germagnoli, Energia Nucleare 5, 591 (1958).

[7] D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. 30, 585 (1958).

[8] See, for instance, D. Turnbull and R. Hoffman, Acta Met. 2, 419 (1954); E. Hart, Acta Met. 5, 597 (1957); A. Mortlock, Acta Met. 8, 132 (1960).

[9] G. V. Kidson, Phil. Mag. 13, 247 (1966).

[10] B. F. Dyson, T. Anthony, and D. Turnbull, J. Appl. Phys. 37, 2370 (1966).

[11] W. Seith and A. Keil, Z. Phys. Chem. B22, 350 (1933).

[12] J. Wesley Miller, Phys. Rev. 181, 1095 (1969); Phys. Rev. B 2, 1624 (1970); and W. K. Warburton, Scripta Met. 7, 105 (1973).

[13] I. E. Leksina and S. I. Novikova, Fiz. Tverd. Tela 5, 1094 (1963) [Sov. Phys. Solid State 5, 798 (1963)].

[14] J. N. Mundy, Phys. Rev. B 3, 2431 (1971).

[15] R. F. Peart and J. Askill, Phys. Status Solidi 23, 263 (1967); G. Neumann, V. Tolle, and W. Hirschwald, Phys. Status Solidi 54, 519 (1972).

[16] J. Wesley Miller, Phys. Rev. 188, 1074 (1969); J. Wesley Miller and W. A. Edelstein, Phys. Rev. 188, 1081 (1969).

[17] A. Seeger, J. Less-Common Metals, 28, 387 (1972); P. S. Ho, Phil. Mag. 26, 1429 (1972).

[18] W. K. Warburton, Phys. Rev. B 7, 1341 (1973).

[19] J. J. Burton and F. Froozan, Phil. Mag. 27, 473 (1973).

[20] D. L. Decker and H. B. Vanfleet, Bull. Am. Phys. Soc. 18, 428 (1973).

[21] T. R. Anthony, B. F. Dyson, and T. Turnbull, J. Appl. Phys. 37, 2925 (1966).

[22] J. A. Weyland, J. Appl. Phys. 42, 5203 (1971).

[23] C. T. Candland and H. B. Vanfleet, Phys. Rev. B 7, 575 (1973); H. B. Vanfleet, D. L. Decker, and C. T. Candland, Bull. Am. Phys. Soc. 18, 428 (1973).

[24] R. A. Johnson and E. Brown, Phys. Rev. 127, 446 (1962).

[25] R. W. Keyes, J. Chem. Phys. 29, 467 (1958).

[26] R. P. Huebener and C. G. Homan, Phys. Rev. 129, 1162 (1963).

[27] R. M. Emrick, Phys. Rev. 122, 1720 (1961).

[28] J. B. Hudson and R. E. Hoffman, Trans. AIME 221, 761 (1961); N. H. Nachtrieb, H. A. Resing, and S. A. Rice, J. Chem. Phys. 31, 135 (1959).