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Modeling and measurement of Al interlayer diffusion in Pd(100): A low-energy ion scattering study

M. J. Gladys, F. Samavat, B. V. King, and D. J. O'Connor

School of Mathematical and Physical Sciences, University of Newcastle, Callaghan, New South Wales, 2308, Australia

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The Pd(001)-$(2\times2)p4g$-Al surface consists of 0.5 monolayer of Al diffusing into the second layer during annealing and causing a $p4g$ clock reconstruction. This is an interesting bimetallic alloy due to the ability to shift the Al cyclically and reproducibly from the second layer to the first and then back again. This is achieved by lifting the Al from the second layer by the absorption of O_2 at room temperature and then removing the oxygen with hydrogen at 200 °C. If the surface is again heated, the Al diffuses back into the second layer and again produces the clock reconstruction. This cyclic process has been found to be repeatable continuously, and scanning tunneling microscope studies have revealed that the diffusion takes place midterrace rather than at a terrace edge. A model has been derived to calculate the activation energy for this process using Fick's second law with a special boundary condition on the second atomic layer. Furthermore, using low-energy ion scattering, the activation energy for the diffusion of Al from the first to the second atomic layer has been measured to be 0.41 ± 0.02 eV. This value suggests that Pd atoms are segregating through the Al to the surface via the exchange process. Measurements at higher temperatures have revealed that the activation barrier for diffusion between the second and third layers is 2.0 ± 0.4 eV, which suggests that the diffusion of the Al into the bulk is via the vacancy mechanism.

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I. INTRODUCTION

AlPd alloys are increasingly utilized in the catalysis industry with applications ranging from exhaust gas purifiers[1] and aerogel catalysts[2] to sulfur extraction.[3] There is also much interest in the growth of Pd/Al alloys by diffusion.[4] The AlPd surface alloy under investigation in this study forms a rather rare surface structure known as a $p4g$ clock reconstruction. The structure consists of a clockwise/anticlockwise rotation of atoms in the unit cell on the topmost atomic layer as shown in Fig. 1. This reconstructed surface has been shown to exhibit unique catalytic properties over bulk AlPd alloy and pure Al or Pd.[5] The surface is generated when Al is deposited on a Pd(100) surface and annealed to temperatures above 700 K. Using low-energy ion scattering (LEIS) Shen *et al.*[6] showed that there was 0.5 monolayer (ML) Al trapped in the second layer as pictured in Fig. 1(a) and no Al at all present on the surface layer. Subsequent low-energy electron diffraction (LEED) measurements revealed a $c(2\times2)$ alloy structure in the second layer. As shown in Fig. 1(b), the top layer of Pd has formed the $p4g$ clock reconstruction. The lateral displacement of the clock reconstruction was measured using LEIS to be 0.5 ± 0.1 Å.[6]

Only recently has the pathway of the Al moving into the second layer been observed. Kishi *et al.*[7] have shown with scanning tunneling microscopy (STM) that the Al diffuses through terraces and not by step edges or special sites, which is the more common mechanism. Further investigations into the growth mechanisms produced contrasting models for the $p4g$ surface structure. Instead of the Al being incorporated into the second layer and mixing with the Pd atoms, it was believed that surface Pd atoms segregate into the through the aluminum adlayer.

As an extension to the AlPd $p4g$ study, Shen *et al.*[6] investigated the consequence of oxygen adsorption onto the $p4g$ surface using LEIS. The oxygen adatoms lifted the 0.5 ML of Al atoms from the second layer to the surface. This process has the effect of removing the $p4g$ reconstruction and producing the original Pd(100) surface beneath a disordered AlO overlayer. The oxygen was removed by exposure to hydrogen at 375 K leaving a Pd(100) surface with 0.5 ML of Al adsorbed on top. If the surface is then reheated to above 700 K, the Al diffuses into the second layer and again produces the clock reconstruction. This cyclic process has been found to be repeatable continuously. Annealing to higher temperatures, 1000 K, causes the Al to diffuse into the bulk, and the surface also resumes the Pd(100) structure.

This paper focuses on the determination of the activation energy for diffusion of Al in Pd(100) using LEIS, by measuring the change in surface concentration of Al, as a function of temperature. The diffusion is modeled using Fick's second law, for nonsteady state diffusion.

LEIS is a necessary tool for this measurement of Al dif-

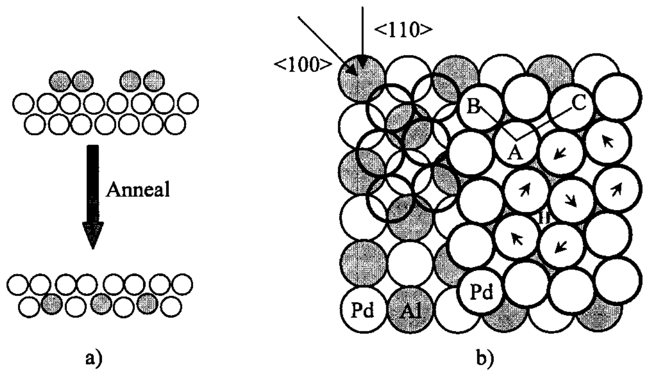

FIG. 1. (a) Process of Al diffusing into the second layer and forming the $c(2\times2)$ structure to maximize mixing. (b) The Pd(001)-$(2\times2)p4g$-Al surface, illustrating the top layer clock rotation caused by the $c(2\times2)$ ordered second layer.

fusion into the second layer due to its ability to measure the concentration of Al on the first and second atomic layers separately.

II. MODELING

Two separate processes need to be modeled. The first is the process of Al diffusion into the second layer over the temperature range of 400–700 K where the Al is trapped and generates the $p4g$ reconstruction. The second process involves the Al that is trapped in the second layer diffusing into the bulk at temperatures above 950 K.

A. Diffusion from first to second layer

Consider a layer-by-layer model for the AlPd system. In this model there is a small energy barrier for the Al to overcome from the first layer to the second layer; however, given that the $p4g$ structure is a more stable structure and that the Al is not observed to jump back to the surface, it is concluded that the energy barrier to jump from the second layer to the first is much greater than that to go from the first to the second layer. The rate of Al atoms diffusing from layer 1 to layer 2, R_{12}, is given by

$$R_{12}=\frac{\partial C_2}{\partial t}=-\frac{\partial C_1}{\partial t}=D_A\left[\frac{\partial^2 C_1}{\partial x^2}\right] \tag{1}$$

for a nonsteady state process where D_A is the diffusion coefficient, and C_1 and C_2 are the concentrations of Al in the first and second layers, respectively. The diffusion coefficient is influenced if vacancies and interstitials alter the diffusion process. Thus, D_A is given by the summation of the three diffusion processes atomic exchange, vacancy, and interstitial, and is given by

$$D_A=F_E D_E C_E+F_V D_V C_V+F_i D_i C_i \tag{2}$$

where F is the probability factor, D is the diffusion coefficient, and C is the local concentration of a given species in units of atomic fraction. Diffusion measurements have shown that typically one of the mechanisms dominates the diffusion process; therefore, if two of the terms in Eq. (2) are negligible then Eq. (1) becomes

$$R_{12}=FA\nu e^{-E_A/kT} \tag{3}$$

with the single measurable activation energy E_A for either exchange, vacancy, or interstitial diffusion, F the probability factor, A the yield parameter for the ion beam, ν the attempt frequency, k Boltzmann's constant, and T the sample temperature.

We need to estimate the probability factor F for the jump, and we do so from the knowledge that the stable end state for the concentration of Al on the second layer (C_2) will be 0.5. If the initial concentration of Al adsorbed onto the Pd surface is 0.5 ML, then the total concentration in both first and second layers is 0.5 ML, i.e.,

$$C_1+C_2=0.5. \tag{4}$$

We assume $C_i=0$, $i>2$. The probability for an Al atom jumping into the second layer is dependent on two factors.

(1) The concentration of Al on the surface (C_1) available to jump.

(2) How far away from an equilibrium end concentration the second layer is, i.e., $0.5-C_2$. The assumption used here is that 0.5 is the lowest-energy state and the most stable state. If there is 0.5 ML, then additional Al (if available) jumping from the first to the second atomic layer will result in a higher-energy state, which is unlikely. Thus there is a term $(0.5-C_2)$, which from the equation above is C_1.

The product of these two terms is normalized to unity by including the term 2 in Eq. (5).
Therefore, the probability factor F is given by

$$F=C_1\times 2(0.5-C_2)=2C_1^2. \tag{5}$$

Thus Eq. (3) becomes

$$R_{12}=-\frac{\partial C_1}{\partial t}=C_1^2 A\sigma\nu e^{-E_A/kT}. \tag{6}$$

Rearranging Eq. (6),

$$-\frac{dC_1}{C_1^2}=A\sigma\nu e^{-E_A/kT}dt, \tag{7}$$

and integrating both sides of Eq. (7),

$$-\int_{0.5}^{C}\frac{dC_1}{C_1^2}=\int_0^t A\sigma\nu e^{-E_A/kT}dt, \tag{8}$$

we get

$$\left[\frac{1}{C_1}\right]_{0.5}^{C}=[tA\sigma\nu e^{-E_A/kT}]_0^t. \tag{9}$$

Therefore, the Al concentration as a function of time is given by

$$C=\frac{1}{2(1+A\sigma\nu e^{-E_A/kT}t)}. \tag{10}$$

Equation (10) is the ideal concentration profile for the AlPd $p4g$ system and can be used to determine the activation energy for the temperature regions observed in the $p4g$ transition by STM, LEED, and LEIS by Kishi *et al.*,[7] Onishi *et al.*,[8] and Shen *et al.*,[6] respectively.

The temperature region where Al is observed to diffuse into the second layer is between 400 and 700 K for LEED and STM. Figure 2 displays surface concentration changes with temperature for different activation energies in the temperature region in which the reconstruction is observed. The scaling parameter A is also adjusted to keep each curve within the 400–700 K region.

Equation (10) can be rearranged so that a plot of $\ln(1/2C-1)$ versus k/T produces a slope equal to the activation energy of the diffusion barrier between the first and second layers. This is an Arrhenius plot and is shown in Fig. 3 for each of the activation energies used in Fig. 2. Any adjustment of the scaling parameter A does not alter the

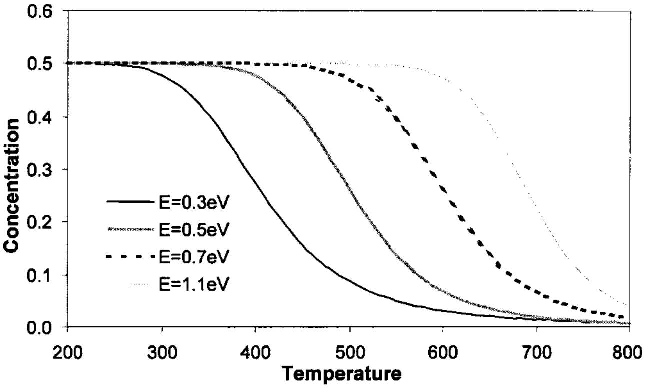

FIG. 2. Plot of Al concentration versus annealing temperature from the model in Eq. (9). The diffusion is concentrated around the region between 400 and 700 K as seen with LEED and STM.

slope of this graph. A comparison of the experimental and calculated results of this graph allows complete determination of the activation energy for the process and an evaluation of the scaling parameter.

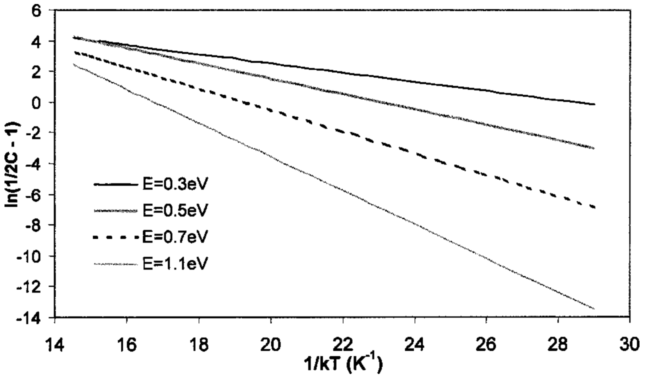

FIG. 3. Plot of $\ln(1/2\,C-1)$ vs $1/T$ from the model of Eq. (9). The slope of the graph is equal to the activation energy for diffusion and the y intercept is given by $\ln(tA\sigma\nu)$.

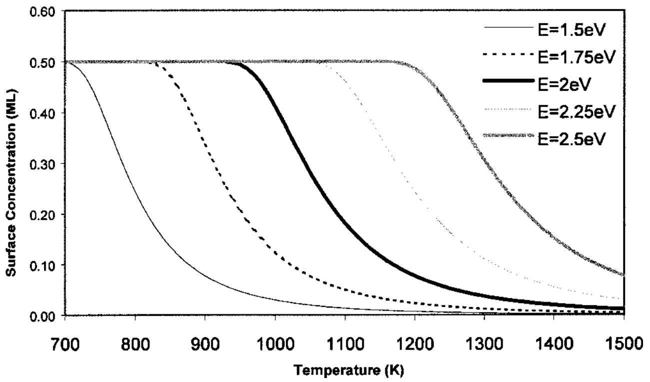

FIG. 4. Concentration versus $1/T$ with interatomic spacing $d=0.389$ nm for activation energies between 1.5 and 2.5 eV and $tD_0=5\times10^8$ nm^2.

B. Diffusion from second layer to bulk

The second process involves the diffusion of Al from the second layer to the bulk. The model derived for the diffusion of 0.5 ML of Al into the second layer of Pd(100) falls short in accurately describing the kinetics for the further diffusion of the Al atoms into the bulk at higher temperatures to that of the $p4g$ transition. This is because the model used a special boundary condition on the second layer. Furthermore, there is a specific interaction between the surface and subsurface layers that increases the effective bulk concentration seen by the surface, and a local equilibrium is reached. The diffusion of the Al from the second layer into the bulk or the diffusion of Pd atoms toward the surface would have a more traditional concentration gradient but would be much slower than predicted by diffusion equations. Du Plessis[9] described this phenomenon as the desegregation rate limited by diffusion. However, the problem is complicated by the initial concentration starting in the second layer. To solve this problem a model must be derived that combines desegregation and diffusion into a semi-infinite solid from a thin planar source as described by Borg and Dienes.[10] If we assume that the rate of decay of the concentration at the surface is identical to that observed if the Al was at the surface (i.e., Al does not jump back to the surface) then from the derivation by du Plessis[9] for a continuous distribution into the bulk, the solution of Fick's second law has the form

$$C=C_0\,\mathrm{erf}\left(\frac{x}{\sqrt{4Dt}}\right). \tag{11}$$

Equation (11) is a solution for a continuous solid, not a discrete set of layers; however the discrete solution can be found by setting $x=d$ (the interatomic spacing). We can then substitute the diffusion coefficient $D=D_0\exp[-E/kT]$ into Eq. (11) to get the relationship between concentration and temperature at the surface as

$$C=C_0\,\mathrm{erf}\left(\frac{d}{\sqrt{4D_0t}}e^{E_A/kT}\right). \tag{12}$$

Figure 4 shows a set of concentration curves for Eq. (12) corresponding to activation energies between 1.5 and 2.5 eV for temperatures higher than 700 K. Figure 5 shows the concentration dependence on D_0t in Eq. (12) for $E_A=2$ eV. The comparison of the changes in E_A (factor of 0.25 from 2 eV)

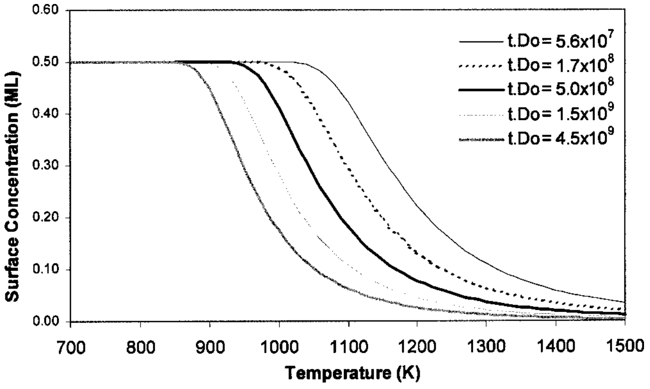

FIG. 5. Concentration versus $1/T$ with activation energy$=2$ eV with tD_0 changed by a factor of 3 from $tD_0=5\times10^8$ nm^2.

and D_0t (factor of 3 from 5×10^8 nm^2) shows how the concentration curve is much more dependent on the activation energy than D_0t.

III. EXPERIMENT

The LEIS experiments were carried out in a Leybold-Heraeus UHV chamber equipped with an angle-resolved ion scattering system. The base pressure in the UHV system is 1×10^{-10} mbar. A 1.9 keV He^+ beam was used at a polar angle of 30° and a scatter angle of 60°. The typical current of the He^+ ions onto the target is in the range of 100 to 400 nA. The scattered particles were analyzed by a electrostatic analyzer ($\Delta E/E=0.005$). High-purity (greater than 99.999%) Al was evaporated from a near-horizontally mounted Al_2O_3 crucible encircled with tungsten wire at 6 A (50 V) with a chamber pressure of 4×10^{-10} Torr. The Al evaporation was approximately 1 ML per 5 min. The measurement of the diffusion of Al into Pd using LEIS has many advantages and disadvantages. Helium ions are the beam of choice due to their low sputter rate as compared with neon or argon. As stated earlier, Al is preferentially sputtered from the surface due to Al being lighter than Pd. The time for analysis is critically important. There is a trade-off between acquiring enough signal to produce good data and minimizing Al sputtering off the surface. Measurements were made to determine the rate of change of Al on the surface due to sputtering by the He ion beam, and the dose (D_i) required to make a perceptible change in Al concentration was determined. In all experiments in this series the total measurement dose was only 10% of D_i to ensure that sputtering had no impact on the change in measured Al concentration. The time to obtain each Al concentration was 15 s.

IV. RESULTS AND DISCUSSION

A. Diffusion from first to second layer

The concentration measurements were performed with an initial coverage of 2–3 ML Al on the Pd(100) surface. However, to prepare a precise amount of Al on the surface, the AlPd $p4g$ cycle was utilized. On heating to 750 K, the Al fills half of the second layer of Pd(100) and produces the $p4g$ reconstruction of the topmost layer of Pd. Any excess Al diffuses further into the bulk. O_2 is then used to lift the half monolayer of Al back to the outermost layer due to the high bonding energy between Al and O. The oxygen is then removed using H_2 at 375 K. This process generates a consistent 0.5 ML of Al on the surface, assuming little sputtering by the ion beam during measurement. This was also compared to the Al and Pd peak heights during dosing as Al grows in layer-by-layer mode. With this knowledge the measurement of the Al peak can be normalized to 0.5 ML for comparison to the Arrhenius plot shown in Fig. 3.

The temperature was increased using radiation heating via a filament up to 750 K and then e-beam heated from 750 to 1150 K. The sample azimuth was rotated to the ⟨100⟩ direction to ensure that only the concentration of Al and Pd on the very topmost atomic layer was measured. An energy spectrum was taken at each temperature increment with energy windows including the Al and Pd peaks.

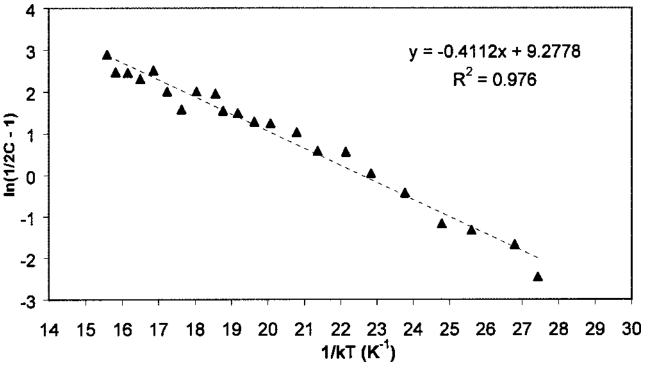

FIG. 6. Arrhenius plot of Al concentration vs $1/kT$. Slope equal to activation energy of 0.41 eV.

An Arrhenius plot for the data taken of Al diffusing into the Pd surface is shown in Fig. 6 of $\ln(1/2C-1)$ vs k/T. The slope in the graph signifies a single diffusion barrier present throughout the temperature region used in the measurement. The slope gives the activation energy for the diffusion barrier and was determined to be 0.41 ± 0.02 eV (41 ±2 kJ/mol). The experiment was reproducible and similar values (within 10%) of the activation energies were measured. There is a very good agreement in Fig. 7 between experiment and Fick's law of diffusion.

The activation energy for diffusion of Al in Pd has never been measured or calculated before, so no comparison can be made with other diffusion experiments. Other systems such as the self-diffusion of Al-Al and Pd-Pd have activation energies of 130 (Refs. 11, 12, 13, and 14) and 266 kJ/mol,[15] respectively. In addition, there are extensive studies on the diffusion of Al in Ni and Fe substrates. The self-diffusion of Ni-Ni has activation energy of 288 ± 10 kJ/mol,[16] while the activation energy for Al impurities in Ni was measured to be slightly less at 274 kJ/mol. This value is in the typical range for vacancy diffusion in Ni, all fcc metals, including Pd, the ratio of Q/RT_m (where Q is the activation energy and T_m is near melting temperature) for self-diffusion is in the range

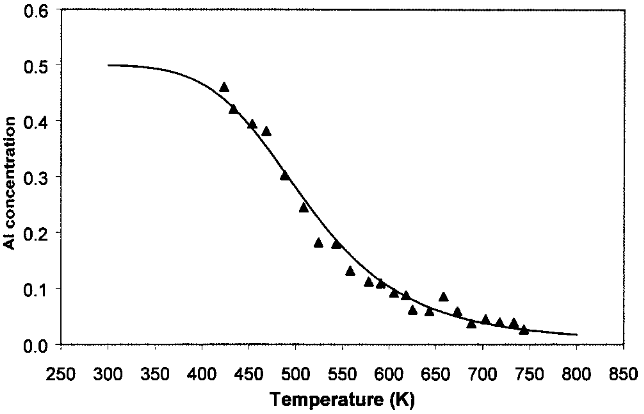

FIG. 7. Comparison of the Al concentration for the model based on Fick's second law of diffusion and the experimental results taken by LEIS.

17–18.[17] Ordinarily impurity diffusion in fcc metals has an activation energy within ±25% of the self-diffusion values[18] and corresponds to diffusion via vacancies. This is also true for the Al in Fe case studied by Taguchi *et al.*[19] However, in contrast to the Al in Ni system, the activation energy for Al in Fe is larger than the self-diffusion activation energy for Fe. For our system, $Q_{(\mathrm{imp})}/Q_{(\mathrm{Pdself})}=0.41/2.6=20\%$, indicating that the diffusion for the first layer to the second is very different from that in the bulk.

Since the activation energy for Al impurity diffusion in the bulk has never been measured or calculated, the bulk activation may be assumed to be the self-diffusion barrier of 266 kJ/mol. Since Pd is a fcc metal the actual value is assumed to be within 25% of this value.[18] Furthermore, the majority of values of the activation energy of Al impurities in other metals such as Be, Au, Pt, Ta, and Ti are located in the lower boundary of the 25% energy range, i.e., in the region that is lower than the self-diffusion value.[16] With this in hand, the activation energy for the Al diffusion via vacancies in the bulk is most likely in the region of 2.0–2.6 eV (210–266 kJ/mol).

The expected activation energy for diffusion of Al in the surface of Pd(100) is different from that in the bulk due to relaxation of the surface layers. The Allen and Wach[20] method calculates the vacancy formation energy at the surface. The barrier for diffusion via vacancies decreases by a factor equal to the ratio between the coordination in the surface and the bulk. For the fcc (100) plane, the surface to bulk coordination ratio equals $\frac{2}{3}$. The diffusion barrier for Al diffusing into the Pd(100) surface via vacancies would therefore have an energy of approximately 1.8 ± 0.2 eV (190 ± 20 kJ/mol). Since the value determined through LEIS is much smaller than this, it may suggest that the process for the diffusion is the exchange mechanism, which at the surface has a lower barrier to diffusion.

The precise determination of the activation energy for vacancy diffusion of 0.7–0.8 eV suggests that the value obtained experimentally of 0.41 eV must not be due to the diffusion of Al in Pd(100) via vacancies. As stated earlier, the self-diffusion energy for Al is 1.3 eV; therefore using the Allen and Wach[13] method for calculating diffusion barriers at the surface, the expected activation energy for diffusion at the surface of Al(100) is 0.8 eV, which is closer to the experimental value but nonetheless still too high in energy. This may suggest that it is not the Al that is diffusing into the Pd second layer but instead the Pd is diffusing out through the Al overlayer. This description agrees with the results of Kishi *et al.*[8] using STM analysis. The palladium located beneath the Al overlayer requires 0.41 eV energy to jump into the layer above. A palladium atom in a subsequent lower layer contributes a Pd atom that diffuses into the second layer, and so on. This process will continue until there is a balance between the segregation energy of Pd and the energy of mixing Al and Pd in the second layer. The growth of Al on Pd(100) is layer by layer; consequently, 0.5 ML of Al on the surface will orient in islands on the surface with local areas having 1 ML coverage. The low value of the activation energy may signify that the process for this diffusion is atom exchange via the ring diffusion mechanism.

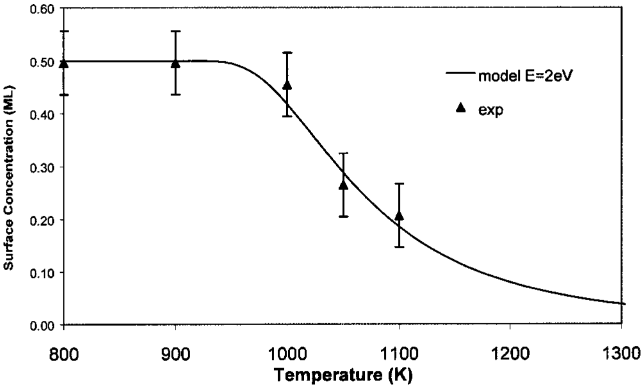

FIG. 8. Comparison of experimental data and Eq. (12) for Al concentration versus sample temperature.

The activation energy of 0.41 eV measured using LEIS may be different from the actual diffusion barrier due to the interaction of the ion beam. For example, induced segregation would lower the apparent diffusion barrier for Al segregating toward the bulk due a flux of defects toward the surface. In addition, enhanced diffusion may also decrease the apparent diffusion for Al to diffuse into the second layer. The use of He as the ion beam and the relatively short measurement time provides confidence that the ion beam effects are minimized and the determined activation energy is within 0.02 eV.

B. Diffusion from second layer to bulk

Measurement at higher temperatures than that forming the $p4g$ sees the Al that is trapped in the second layer now available to diffuse into the bulk. The second layer concentration is measured by rotating the sample azimuth to a position in which the second layer is no longer shadowed by the first, i.e., a few degrees off the ⟨110⟩ direction as shown in Fig. 1. The concentration of the second layer Al is obtainable since the first layer is pure Pd and the second is 50% Al and 50% Pd. Therefore scattering from the first and second layers produces peaks of Al (layer 2) and Pd (layers 1 and 2). Figure 8 shows the results from the measurement of second layer concentration versus annealing temperature, compared to the concentration model from Eq. (12).

The measurement of the second layer concentration as a function of annealing temperature had several complications. First, the intensity of the Al peak is very low due to the Al sitting in the second layer as well as the Pd surface layer transformed into the clock reconstructed $p4g$ surface. In addition to this, the annealing temperature needed for this experiment necessitated the use of *e*-beam heating. The high voltage on the filament needed for electron acceleration caused distortions in the He ion beam used in the measurement. These fluctuations resulted in fluctuations in the counts by the analyzer and limited the experiment to only a few measurements. This is also the origin for the large error bars attached to each measurement in Fig. 8.

The potential energy diagram for the system is shown in Fig. 9, and has the activation energy for vacancy diffusion of Al in bulk Pd calculated to be 2.1 eV. $\Delta E_{1\text{-}2}$ is the measured

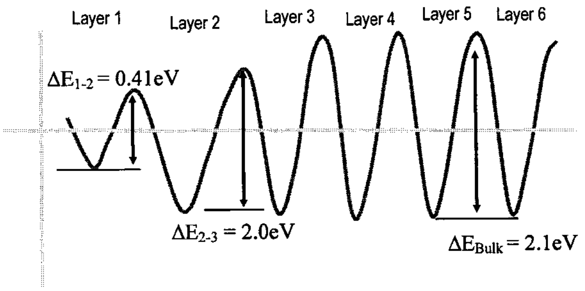

FIG. 9. Energy potential diagram for diffusion of Pd from second layer through Al overlayer onto surface.

activation energy (0.41 eV) for Al diffusing from the surface and into the second layer. $\Delta E_{2\text{-}3}$ is the activation energy for Al to overcome to move into the third layer and is equal to 2.0 eV. The activation energy for the diffusion of Al from the second layer to the first is an unknown quantity. It may be equal to, less than, or even greater than $\Delta E_{2\text{-}3}$. If $\Delta E_{2\text{-}1} = \Delta E_{2\text{-}3}$ the probabilities for a jump to occur to either the first or third layer are equal. The probability for an Al atom in the first layer to jump into the second layer is close to unity, whereas there is approximately an equal probability for an Al atom to jump from the third layer to either the second or fourth. Consequently, the Al trapped in the second layer will form a concentration gradient into the bulk, similar to a traditional Fick's second law problem (i.e., no special boundary conditions).

The activation energy determined for diffusion from the second layer to the bulk is very close to the value obtained from the analysis derived earlier of Al impurity diffusion through the bulk (2.1 eV). This suggests that the Al is diffusing through the Pd via vacancies.

V. CONCLUSION

Analysis of the transition from Al/Pd(100) to Pd(100)-Al-$(2\times 2)p4g$ over the temperature range 400–700 K and the diffusion of Al into the bulk of Pd(100) at temperatures higher than 1000 K has indicated two distinctly different diffusion processes. The apparent transition of Al into the second layer of Pd to form the $p4g$ structure was found to have an activation barrier of 0.41 ± 0.02 eV. This may suggest that Pd is segregating through the surface layer of Al, not by Al diffusion into the second layer. The exchange mechanism is the most likely candidate for the observed diffusion process. The second layer Al atoms were found to diffuse further into the bulk at temperatures higher than 1000 K. The diffusion barrier for this process was determined to be 2.0 ± 0.4 eV, which suggests diffusion of the Al through Pd via vacancies.

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