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Positron-lifetime studies of physisorbed nitrogen and argon surfaces

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Positron-annihilation lifetimes have been measured in two-dimensional nitrogen and argon adsorbed on Grafoil surfaces as a function of coverage and of temperature. Two positron lifetimes were observed, a short lifetime ≈ 0.2 nsec which is attributed to annihilation in the bulk of graphite and a long-lived component ≈ 0.4 nsec which is attributed to annihilation in adsorbates and on the graphite surface. The surface lifetime increases as a function of adsorbate coverage, reaching a maximum near half a monolayer and then decreasing. This lifetime variation with the fraction of coverage is interpreted to be a result of the positron specifically annihilating with the surface defects created by the presence of adsorbates. The positron lifetimes were found to vary with temperature for surfaces with a fixed fraction of monolayer adsorption. The temperatures at which the significant lifetime changes occur were found to correspond to the known solid-liquid phase-transition temperatures for N_2 and Ar two-dimensional surfaces. The lifetime-temperature variation was found to be weaker for two-dimensional adsorbates than in three-dimensional solid-liquid phase transitions.

I. INTRODUCTION

In recent years there has been a growing interest in the study of the adsorbed atoms and molecules which interact by the relatively weak van der Waals forces with each other and with the substrates on the surfaces where they reside. The condensed "physisorbed" states of adsorbates on a two-dimensional $(2D)$ surface,¹ analogous to the bulk states, constitute a simpler system for fundamental studies. Due to the lack of strong long-range ordering, studies of 2D systems have given much detailed quantitative information about various fundamental problems² such as melting, wetting, and microstructures on surfaces. The importance of 2D systems is also great in studying phenomena which have applications, such as thin films, metallic and magnetic layer compounds, inversion layers of semiconductors, smectic liquid crystals, and dragging in hydrodynamics.²

In recent years research on 2D systems has been pursued both theoretically and experimentally. Theoretical approaches using both quantum and statistical mechanics have had great success in interpreting experimental phenomena. Experimental investigations on 2D systems have been made using various experimental techniques, such as low-energy electron diffraction (LEED), photoemission (PE), Auger electron spectroscopy (AES), x-ray and neutron diffraction, nuclear magnetic resonance (NMR), 'heat-capacity measurements, etc.^{1,2} Each technique has a different degree of sensitivity and surface specificity for certain problems. Gn the other hand, each technique has

its limitations, mainly due to the perturbation of the system by the various probes.

The positron,⁴ the antiparticle of the electron, is a newly discovered surface probe. It possesses several unique properties for surface characterization. The work function of a positron has been found⁵ to be negative in many solids. Recent slow-positron results have shown^{4,6} that positrons are preferentially attracted to the surfaces and the resulting positron-annihilation signals are thus enhanced for surface states. Current applications of slow positrons to condensed matter physics has been primarily on the electronic structure of surfaces and interfacial phenomena.^{7,8} However, this research requires ultrahighvacuum conditions and clean surfaces which are different from applied systems in the real world. On the other hand, one can also use fast positrons from radioactive decay in a conventional manner for surface studies. In this case, the systems studied must have high surface areas, such as do fine powders, where the positron signals from the surface are enhanced. Results thus obtained in this manner are more similar to real surface systems and thus complement the information obtained by the slow positron method.

Surface phenomena on graphite and carbon powders are interesting to study by means of fast-positron techniques because the oxidation products of carbon (CO) and $CO₂$) are volatile. Unlike other substrates which give oxide surfaces, one can obtain a clean surface by evacuating and baking the graphite powders for surface studies. Recently, we have reported a series of positron-lifetime mea-

surements by using fast positrons on various graphite surfaces and have found that about 50% of the positron signals can be attributed to the surfaces, as evidenced by the fact that the surface lifetimes are twice those in the bulk. In this paper we wish to report the positron-lifetime results for two well-known 2D systems, i.e., nitrogen and argon on graphite as a function of adsorbate coverage and of temperature.

II. EXPERIMENTAL

A. Sample preparations

The substrate was Grafoil (from Union Carbide Co.) which is an exfoliated recompressed sheet made from graphite particles. The density was measured to be 1.11 $g/cm³$ and the surface area was determined to be 21.2 m^2/g by the nitrogen-adsorption method to be described below. The Grafoil sample was spectro-grade purity, similar to those used in neutron-diffraction studies.^{10} The Grafoil (26.9 g) was baked at 900 °C under vacuum (10^{-6}) torr) for 48 h in order to remove all impurities. The baked sample was then transferred to a sample cell made of aluminum in a well-sealed glovebox under a dry atmosphere of high-purity helium. The positron source was embedded in the center of the packed Grafoil sample. The positron source was supported on a thin Ni foil (0.85) $mg/cm²$). The sample cell was placed in a closed-cycle He refrigerator (Air Product Displex). The sample was evacuated under vacuum (10^{-6} torr) for 10 h before measurements were made. The positron-lifetime result on the clean Grafoil samples was found to be the same as our previous result⁹ for a sample which was post-baked in situ at 600° C for 24 h. The bulk N₂ and Ar phases were condensed directly from research-grade gases near the boiling points in a separate cell (5 cm^3) in the center of which the positron source was located.

B. Adsorption of N_2 and Ar on Grafoil

The adsorption of N_2 and Ar and the selection of a fractional coverage on the Grafoil surfaces were done using the isotherm method.² N_2 and Ar gases (research grade) were obtained from Matheson Gas Co. (Secaucus, NJ). They were introduced into the cell through a gas manifold consisting of a standard volume (1000 cm^3) and two absolute pressure gauges (a MKS transducer and a Veeco ion gauge). The design of the gas-handling system is similar to those reported in neutron-diffraction experiments.¹⁰

The adsorbed volume-pressure isotherms obtained for N_2 and Ar are shown in Figs. 1 and 2. The extrapolated monolayer capacities between the lines at the Henry's-law region (low coverage) and the lines from the plateau were found to be 138 ± 1.0 cm³ and 135 ± 1.0 cm³ at standard temperature and pressure (STP) for N_2 and Ar, respectively. From this amount of gas and by using the N_2 moleculy. From this amount of gas and by using the N_2 molecular surface area (15.7 A^2),¹¹ we obtained a specific surface area for Grafoil of 21.2 m²/g, which is in good agreement with other investigations.^{10,11} The fraction of surface coverage below a monolayer can thus be calculated from the ratio of the adsorbed volume and the monolayer capacity.

FIG. 1. An isothermal plot of N_2 on Grafoil at 77 K. The dashed lines are extrapolated from the low-coverage and the complete-monolayer regions. The adsorbed volume $(=138 \text{ cm}^3)$ at the intercept corresponds to the monolayer capacity (see the text). The solid line is a smoothed curve drawn through the data points.

In the case of constant-coverage experiments, the fractional coverage was first obtained by the isotherm technique at 77 K. The cell (dead volume equal to 12.5 cm^3) pressure was monitored and changed by adjusting the gas vapor pressure while decreasing the temperature from $77-10$ K in order to maintain a fixed quantity of adsorbate on the surface. The vapor-pressure adjustments were based on the logarithmic pressure and the $1/T$ relationship from the known volume of the system, the measured vapor pressure, and the cell temperature. A typical length of time in preparing a surface coverage at a certain temperature was about 2 h. All temperature measurements were made with calibrated germanium and platinumresistance thermometers and a Au-Fe-Cr thermocouple

FIG. 2. An isothermal plot of Ar on Grafoil at 77 K. The dashed lines are extrapolated from the low coverage and the complete monolayer regions. The adsorbed volume $(=135 \text{ cm}^3)$ at the intercept corresponds to the monolayer capacity (see the text). The solid line is a smoothed curve drawn through the data points.

embedded in three different positions in the Al cell and at the cold tip of the cryostat. The cell temperature was kept at a gradient less than ± 0.1 K across the cell for each lifetime experiment.

The positron-lifetime measurements were carried out using a conventional fast-fast coincident method by monitoring the starting signal (1.28 MeV γ ray) from positron decay in 22 Na isotopes and the stopping signal (0.51 MeV γ ray) from the positron annihilation in the studying material. The resolution of the spectrometer was found to be about 300 psec by measuring the coincident photons from a ⁶⁰Co source. The lifetime spectra were fitted with a sum of negative exponential terms by a computer program (PO-SITRONFIT EXTENDED). The actual resolution for the spectra was represented by two Gaussian functions. The lifetimes were carefully fitted using three components, where the third component had a fixed lifetime $\tau_3 = 1.3$ nsec and intensity $I_3=1\%$ due to the o-Ps contribution from the Grafoil surface as reported in our previous paper.⁹ A source correction due to the Ni foils was also made in the computer analysis. Results of the two lifetime components, $\tau_1 \approx 0.2$ nsec and $\tau_2 \approx 0.4$ nsec are reported in this paper for detailed discussions. The errors shown in the table and figures are the statistical standard deviations from the computer-fitted results.

C. Positron-lifetime measurements III. RESULTS AND DISCUSSIONS

A. Positron lifetimes in the bulk of N_2 and Ar

It is known that the positron-lifetime changes in a solid-liquid phase transition in bulk systems [threedimensional (3D)].⁴ It is important for us to report the lifetimes of N_2 and Ar in the bulk (3D) before we study 2D phase transitions. The bulk lifetime results for N_2 and Ar over a wide range of temperatures are shown in Table I.

The lifetime spectra in the bulk were fitted with three lifetimes. A short lifetime, $\tau_1 = 0.125$ nsec, was fixed in the data analysis with a constraint that its intensity equaled one-third the intensity of the long-lived component. The long-lived component, $\tau_3 = 2 - 4$ nsec is easily identified as 0-Ps in the bulk. Therefore we assign the second component to the contribution from the positrons in the bulk. The positron lifetimes obtained, 0.34—0.⁴⁵

^aThe experimental error temperature measurements were ± 0.1 K.

^bThese data have been plotted in Fig. 3.

FIG. 3. Positron lifetimes in the bulk for nitrogen and argon. T_m and T_b represent the melting and the boiling temperatures. The lines were drawn through the experimental data as a guide to the eye.

nsec agree with the results of reported lifetimes for N_2 (Ref. 12) and for Ar (Refs. 13 and 14) (see Fig. 3). The long-lived o-Ps components are due to voids or defects in the solids and to bubble formation in the liquids. Both τ_2 and τ_3 increase as a function of temperature due to the effect of bulk-volume expansion. The values of τ_2 , i.e., the 3D positron lifetime, are important information for us to compare with 2D lifetimes on the surfaces in the following sections.

B. Positron lifetimes on 2D surfaces

We have measured the positron lifetime as a function of coverage for N_2 and Ar adsorbates on Grafoils samples at 77 K. The results of the surface positron lifetimes and the corresponding intensities are shown in Figs. ⁴—6. Since the positron lifetimes in bulk Grafoil have been found to be nearly constant, $\tau_1 = 0.220 \pm 0.009$ nsec, we shall, therefore, only discuss the variation of the surface positron components τ_2 and I_2 with the quantity of adsor-

FIG. 5. Surface positron lifetimes vs fractional Ar coverage on Grafoil at 77 K. The solid line is a fitted curve according to Eqs. (1) and (2). The dashed line is drawn between the lifetimes at the clean Grafoil surface and in the solid Ar (3D).

bate on the surface.

The variations of τ_2 with the fractional coverage shown in Figs. 4 and 5 are interesting in many respects. First, it is interesting to observe a change of τ_2 for this small submonolayer coverage. The ratio of the total amount of adsorbate for a monolayer to the total quantity of Grafoil was found to be only 0.641% and 0.895% for N_2 and Ar, respectively. As shown in Figs. 4 and 5, we start to observe a significant increase of τ_2 at about 0.2 coverage of monolayer, which corresponds to about 0.¹ to 0.2 weight by percent. The sensitivity of τ_2 in response to the adsorption is associated with the surface specificity of the positron. The positron is different from an electron but it usually possesses a negative work function in solids. The positron, therefore, is preferentially diffusing to the surface. The positron work function in graphite has not been reported, but our lifetime results indicate that its work function may also be negative in view of the large fraction of surface positrons observed in Grafoil samples.

FIG. 4. Surface positron lifetimes vs fractional N_2 coverage on Grafoil at 77 K. The solid line is a fitted curve according to Eqs. (1) and (2). The dashed line is drawn between the lifetimes at the clean Grafoil surface and in the solid N_2 (3D).

The variation of τ_2 with coverage shown in Figs. 4 and 5 is also interesting. It reaches a maximum value at about

FIG. 6. Surface positron intensities vs fractional N_2 or of Ar coverage on Grafoil at 77 K.

half a monolayer and then falls back to values near that of the graphite surface or of the bulk adsorbate. The dashed lines in Figs. 4 and 5 link the lifetimes for clean graphite surfaces and the bulk adsorbates. In the absence of surface effects, the dashed line is probably the expected variation. The net difference between the observed lifetimes and the expected lifetimes in Figs. 4 and 5 must be associated with changes in the surface structure or of positron behavior due to the presence of adsorbates.

The adsorption of N_2 and Ar forms a two-dimensional surface through a mechanism called "island formation".¹⁵ The structure of N_2 is a registered commensurate $(\sqrt{3}\times\sqrt{3})R$ 30 phase, ¹⁶ while Ar forms an incommensurate phase¹⁷ due to the different sizes of adsorbates with respect to the graphite structure. Despite this distinct difference in the structures of the adsorbed surfaces, we observe a similar variation in the region of submonolayer adsorption. Since the positron work functions of Ar and N_2 have not been measured, we first discuss the variation of τ_2 as a result of the surface structural changes due to adsorption. In any case, any change of positron behavior, such as in the work function, may be.difficult to resolve by the current technique because the bulk lifetimes of Ar and N_2 are found to be not significantly different from those of the graphite surfaces.

Despite the fact that the clean surface of graphite may not be perfectly two-dimensional, the adsorption must have created some kind of additional imperfection structures on the surface. At a surface coverage below half a monolayer, the quantity of the additional inhomogeneous surface is proportional to the quantity of the adsorption. If we consider this newly created inhomogeneous surface due to the presence of adsorbate as defect centers for the surface positrons to interact with, we expect that the positron lifetime will increase with the fraction of coverage or the quantity of adsorption. We can therefore fit the variation of τ_2 as a function of the monolayer fraction Θ below half coverage as

$$
\tau_2 = \tau_2^0 + k\Theta, \ \Theta = 0 \text{ to } 0.5 \ , \tag{1}
$$

where τ_2^0 is the mean value of the positron lifetime on the clean Grafoil surface and of the bulk adsorbates, i.e., the value of the dashed lines in Figs. 4 and 5, and where k is a fitting parameter which is related to the difference in annihilation rate between adsorbates and graphite surfaces. The maximum value of τ_2 is expected to be at $\Theta = 0.5$ because above $\frac{1}{2}$ coverage the surface becomes more uniform again as more adsorbates are added. For $\Theta > 0.5$, we fit the variation of τ_2 with the fractional coverage by:

$$
\tau_2 = \tau_2^0 + k' \Theta(1 - \Theta), \ \ 0.5 < \Theta < 1.0 \tag{2}
$$

As shown by the solid lines of Figs. 4 and 5, we found that the data could be fit well with Eq. (2) above $\Theta = 0.5$ but less well with Eq. (1) below Θ =0.5. The fluctuation of τ_2 below Θ =0.5 may be due to the fact that the surface of graphite is not perfectly two dimensional and that the adsorbates start by filling the imperfections at very low coverage. This can be seen in Figs. 4 and 5, that τ_2 does not vary significantly below $\Theta = 0.2$ due to the filling of these imperfection sites.

It is also interesting to observe that the maximum change of τ_2 (at half coverage) is larger for N₂ than for Ar. This is probably due to the fact that the positron lifetime in bulk Ar differs more from that of graphite than does the positron lifetime in bulk N_2 (see the dashed lines of Figs. 4 and 5). Another interesting result in Figs. 4 and 5 is that the positron lifetimes for monolayers of Ar and N_2 , i.e., 0.367 nsec and 0.375 nsec, are closer to the values for 3D solids than for 3D liquids. This suggests that the so-called fluid phase at 77 K on graphite surface is behaving more like a solid than a liquid for both $2D N_2$ and Ar surfaces.

The results of the surface-positron intensities I_2 are shown in Fig. 6 as a function of coverage for Ar and N_2 . Contrary to the variation of τ_2 , we observe almost constant values of I_2 with respect to the fractional coverage. The increasing trend of I_2 above one monolayer as shown in Fig. 6 is probably due to the formation of an overlayer which is more similar to the bulk states of N_2 and Ar which contain about 90% of that positron component (Table I). The insensitivity of I_2 with respect to Θ is probably due to the small ratio of adsorbate to Grafoil. Using these results for I_2 , τ_1 and τ_2 , we were not able to fit the two-stage trapping model, which was developed for positron solid-state physics,⁴ for the current surfacedefects systems. This may be due to the change of positron behavior on the surface in the presence of adsorbates, which we have no information on at the moment. The difference between τ_2 and τ_2^0 is too small to be resolved into two states by the current technique. We hope that quantitative studies using a slow positron beam can help to resolve this problem in the future.

C. Positron lifetime and temperature dependence on 2D surfaces

We have measured the positron lifetimes as a function of temperature for constant N_2 and Ar coverages on the graphite surface. The fractional coverages were chosen to be 0.5 and 0.7 monolayer for N_2 and Ar, respectively. The results for τ_1 (=0.22 nsec) were found to be nearly independent of temperature between 15 and 85 K. These τ_1 values are essentially the same as for clean graphite, but τ_2 and I_2 vary as a function of temperature for the adsorbed surfaces as shown in Figs. $7-10$. On the other hand, we found that the surface positron lifetimes and their intensities are nearly independent of temperature as shown in the dashed lines of Figs. ⁷—10. We, therefore, discuss the temperature variations in terms of structural changes on the adsorbed surfaces.

Positron lifetimes are known to be sensitive to various phase transitions, such as solid-liquid,⁴ liquid crystal-
ine,¹⁸ micellar formations,¹⁹ etc. All of these existing studies have been reported for bulk transitions. The structures and related phase transformations on 2D adsorbates have been studied using several techniques, such as LEED, neutron diffraction, and specific heat and isotherm methods. The results in Figs. ⁷—¹⁰ are the first reported positron studies on 2D systems. Our first interest is to discuss these results in the light of 2D phase transformations. The most significant phase transition in

FIG. 7. Surface positron lifetimes for N_2 at 0.5 monolayer coverage on Grafoil as a function of temperature. The solid line is a smoothed curve drawn through the experimental data. The dashed line represents the smoothed lifetimes from the results for clean Grafoil.

2D systems is the solid-liquid transformation (melting). The mechanism of this transformation is of current interest, in particular, the order of the phase transition is of interest.²⁰ In the case of 0.5 monolayer N_2 coverage shown in Figs. 7 and 8, we observe a change in the surface positron lifetime τ_2 at 42 K, which corresponds to the known 2D solid-liquid transition temperature.¹⁵ The sensitivity of the positron lifetime to the 2D phase transition is easily seen in the figure. The τ_2 variation in a relatively narrow temperature range suggests that the melting mechanism is first order, which agrees with the results obtained with other techniques.²¹ In general, a first-order phase transition gives a step-function-type change in the positron lifetimes for 3D systems.⁴ However, if we compare the curve of Fig. 7 with the τ_2 change at the 3D melting temperature of Fig. 3, we find that the increase in the positron lifetime in 2D is weaker and smaller than that in 3D. This difference may result from the fundamental

FIG. 8. Surface positron intensities for N_2 at 0.5 monolayer coverage on Grafoil as a function of temperature. The dashed line represents the smoothed lifetimes from the results for clean Grafoil.

FIG. 9. Surface positron lifetimes for Ar at 0.7 monolayer coverage on Grafoil as a function of temperature. The solid line is a smoothed curve drawn through the experimental data. The dashed line represents the smoothed lifetimes from the results for clean Grafoil.

difference between 2D and 3D systems, such as the lack of long-range interaction involved in the melting mechanism. Another possible reason is that the small weight ratio of 2D adsorbate to the bulk Grafoil has attenuated the observable changes of lifetime at the solid-liquid transition. In any case, this small fraction ($\sim 0.5\%$) of 2D surface is sufficient to give a clear change in the surface lifetime to detect the melting. On the other hand, I_2 (Figs. 8) and 10) does not respond to the 2D melting as clearly as does τ_2 .

For 0.7 monolayer Ar coverage, the results for τ_2 and I_2 are shown in Figs. 9 and 10. We observe a gradual decrease of τ_2 over a wide range of temperature, $\approx 50-70$ K, as shown in Fig. 9. The temperature where τ_2 starts to decrease, $T = 52$ K, agrees with the known melting point for 2D Ar surfaces.¹⁷ The results of Fig. 10 show that I_2 decreases at temperatures between ¹⁷—⁴⁵ K and then increases above the melting point. These results are very different from the results for N_2 systems reported above.

A slower variation of τ_2 in Ar with respect to melting

FIG. 10. Surface positron intensities for Ar at 0.7 monolayer coverage on Grafoil as a function of temperature. The dashed line represents the smoothed lifetimes from the results for clean Grafoil.

indicates that the melting mechanism for 2D Ar is different from that for 2D N₂, where τ_2 shows a sharp variation at the melting temperature (Fig. 7). It is possible that the transition for Ar may be of weaker first order²² than the first-order transition reported for N_2 . Unfortunately, in both cases, our experimental deviations are bigger than the differences in τ_2 or I_2 , possibly as a result of hysteresis due to the temperature effect in the current technique. The difference in I_2 at low temperature between Ar and N_2 may be related to structural differences; N_2 is in a registered $(\sqrt{3}\times\sqrt{3})R$ 30 structure while Ar is in an incommensurate phase.²¹ Another possible reason is that the orientational order-disorder phase transition in N_2 complicates the detectable behavior of I_2 . These various phase transitions on 2D surfaces may be observable by using a monoenergetic positron beam which can probe the surface adsorbate with less interferences from the substrate.

IV. CONCLUSIONS

We have reported positron-lifetime measurements in a 2D system for the first time as a function of coverage and temperature. The positron-lifetime changes as a function of the fractional monolayer coverage. This variation is interpreted as arising from surface imperfections created by

the adsorption. The change of the positron lifetime in such a 2D adsorbed graphite system is sensitive, to the level of 0.1% by weight, to the ratio of the adsorbate and the Grafoil. The positron-lifetime changes at the melting transition on 2D surfaces and gives qualitative information about the order of the transition. The lifetime change is sharper in 2D N_2 than in 2D Ar. Positron-lifetime results do not show responses to the known orientational order-disorder nor to the commensurate-incommensurate 2D phase transitions. Obtaining detailed quantitative information about surface processes —such as phase transitions, the wetting process, mechanisms and orders of transitions, and surface fine structure—by using positron probes appears to be possible in the near future by employing monoenergetic slow-positron sources to observe other positron-annihilation features, such as Dopplerbroadened energy spectra and angular correlations due to positron-annihilation radiation.

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