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Sticking coefficient of nitrogen on solid N_2 at low temperatures

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Abstract

The sticking coefficient of nitrogen gas on a thick solid nitrogen film on a copper cold finger was studied at low temperature. For surface temperatures of about 12 K the sticking coefficient is measured to be $99.0 \pm 0.6\%$. Our result implies that it will be possible to make a intense and high brightness slow positron source starting from a small diameter deposit of the gaseous positron emitter $^{13}N_2$ produced in the reaction $^{12}C(d,n)^{13}N$.

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Keywords: Sticking coefficient; Nitrogen

1. Introduction

An accelerator-produced source of a short-lived positron emitting isotope [1] could be a convenient way of obtaining a high intensity slow positron beam [2]. Several previous efforts have made slow positron sources using the $^{12}C(d,n)^{13}N$ reaction [3–5]. We are presently constructing a slow positron source in which an intense point-like radioactive source of ^{13}N will be formed by extracting gaseous $^{13}N_2$ from a graphite production target and condensing it onto a small area [6] on a thin foil behind a solid rare gas moderator cone [7]. To understand the efficiency of the formation of the solid N_2 deposit from the vapor and to know the proper conditions for obtaining the small spot size required for a high brightness source, we need to know the sticking coefficient of N_2 gas molecules on a solid N_2 surface. (By ''sticking coefficient'', also known as the accommodation coefficient, we mean the probability that a nitrogen molecule impinging upon a surface will afterward be stuck to the surface.) For this purpose, we have studied the adsorption of nitrogen gas as a thick layer on a metal surface at temperatures such that the vapor pressure of N_2 will ensure that the deposit, once formed, evaporates slowly compared with the 10 min half life of ^{13}N.

Because of the interest in molecular interactions with surfaces, the adsorption of nitrogen gas molecules on metal surfaces such as W, Be, Ru, Fe, Ni, Re, etc. has been extensively investigated. To elucidate the nature of the metal atom–nitrogen molecule potential, most of these researches [8–13] have focused on the first monolayer of adsorbed N_2 at room temperature or higher temperature. While there are also experiments reporting on the adsorption of nitrogen on metal surfaces at low temperature [14–16], the adsorption mechanism has been investigated at clean surfaces with relatively low exposures (less than 10 Langmuir) of N_2 gas. After the first few monolayers of nitrogen have been chemi- or physic-sorbed on a surface, further condensation will occur on top of an essentially solid nitrogen surface and form a solid α-nitrogen film, no matter what the substrate is [17,18]. While yielding useful information about the structure of the nitrogen overlayers and the interaction potentials, these studies have not provided us with a value for the sticking coefficient of nitrogen on a previously formed nitrogen film. The purpose of the present communication is to provide this desired information.

2. Experimental

Our measurements have been conducted in a UHV chamber pumped by a 50 l/s turbo pump to a base pressure 6.5×10^{-9} Torr in the absence of cryocooling. The chamber is equipped with a continuous flow liquid He cold finger and a

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manipulator. A nozzle with ID 0.89 mm connected to a leak valve admits nitrogen gas into the chamber and can be positioned, using the manipulator, either directly above the cold surface or pointing toward the vacuum pump. The nitrogen gas is from a lecture bottle labeled 99.9995% pure. A 19 mm diameter OFHC (oxygen-free high conductivity) copper rod was connected to the cold finger and its flat end is used as the substrate for nitrogen adsorption. A temperature sensor was mounted at the side of the rod and very close to the end so that the temperature measured is close to the temperature at the flat surface. The cold finger is surrounded by a hollow cylindrical aluminum heat shield with a 9 mm diameter opening in the face of the cylinder, which is covering the end of the copper rod. The opening is approximately 5 mm from the end of the copper rod. With this arrangement the thermometer on the copper rod reads 12 K with a liquid helium flow rate of approximately 10 l/h.

3. Results and discussion

After the cold finger attained a low temperature (~12 K) the chamber pressure fell by an order of magnitude to a value 7×10^{-10} Torr. N_2 gas was then admitted through the leak valve with the nozzle located outside of the heat shield of the cold finger and the chamber pressure was recorded as P_1. The nozzle was then moved to the front of the cold finger about 0.5 mm away from the surface and the pressure dropped to a much lower value (recorded as P_2) due to the pumping effect of the cold surface absorbing the gas directly after it leaves the nozzle. After 1 min, the nozzle was moved back to the original position in order to prevent the cold finger from being exposed to the nitrogen gas for a long time and so causing the background pressure to rise unduly. In this experiment, a thick nitrogen film is being formed so that chemi- and physic-sorption effects associated with the dirty metal surface are irrelevant after the first few seconds. Thus our estimate of the sticking coefficient refers to gas phase N_2 sticking to a solid N_2 surface.

The measurements were done both at low temperature and room temperature. At room temperature, the sticking coefficients are expected to be much less than unity because probably only one monolayer or two can stick on a dirty metal surface. One would thus expect that P_1 and P_2 should be almost the same, but it is possible that when the nozzle is moved to different positions, the two pressures could be different because of a different effective pumping speed due to the different geometry. In fact, our experiment shows these two pressures are the same to within 2% so that there is evidently no significant pumping speed correction that should be applied to the low temperature measurements.

On the contrary, at low temperature, one might expect the sticking coefficients to be close to unity since there is nothing to prevent more layers from condensing on top of any layer of nitrogen. The experiment verified this assumption and a typical record of pressure measurements as a function of time is given in Fig. 1.

In Fig. 1, at the points labeled A the gas nozzle is outside the shield, far from the cold finger, and the equilibrium gas pressure is P_1. Just after the point A, the nozzle is moved to the front of the cold finger. One minute later at point B the pressure has fallen to P_2. The nozzle was then moved to the outside of the shield again. At point C the leak valve is being closed. P_0 is the background pressure obtained by a three exponential fit to the data beginning at point C. The top and bottom dashed lines are P_1 and P_0, respectively. Since the initial pressure was almost constant, the rapid pressure change after the nozzle was moved to the front of the cold finger is assumed to be caused only by the surface sticking of nitrogen gas on cold surface. The sticking coefficient S may be estimated from the simple expression.

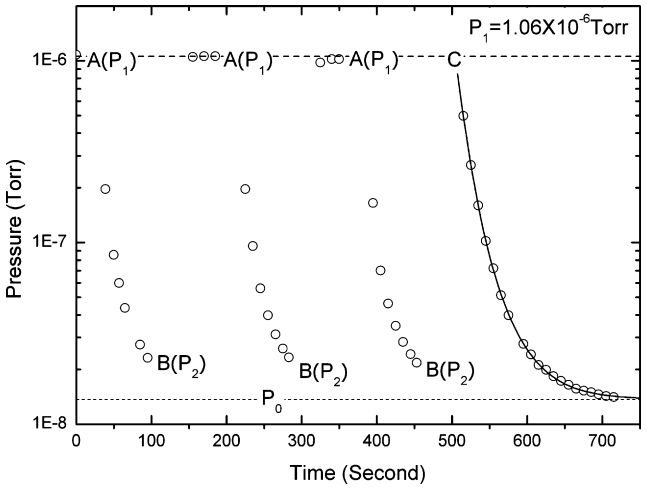

Fig. 1. Typical measurement of pressure as a function of time. At A, the nozzle was moved to the front of the cold finger, and at B, the nozzle was moved to the outside of the shield. At C, the leak valve is being closed.

$$S = 1 - \frac{P_2}{P_1}$$

This expression neglects both the background pressure and the fact that the pressure has not stopped decreasing after 1 min. By removing the effect of the background pressure P_0, the

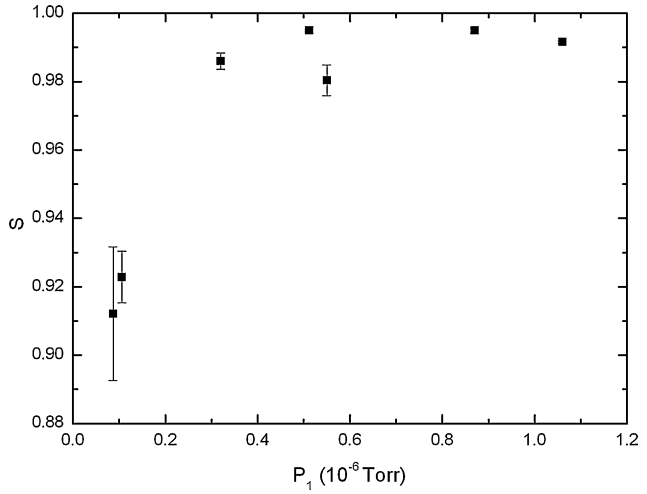

Fig. 2. Sticking coefficient S of N_2 gas on a 12 K surface as a function of N_2 gas pressure P_1.

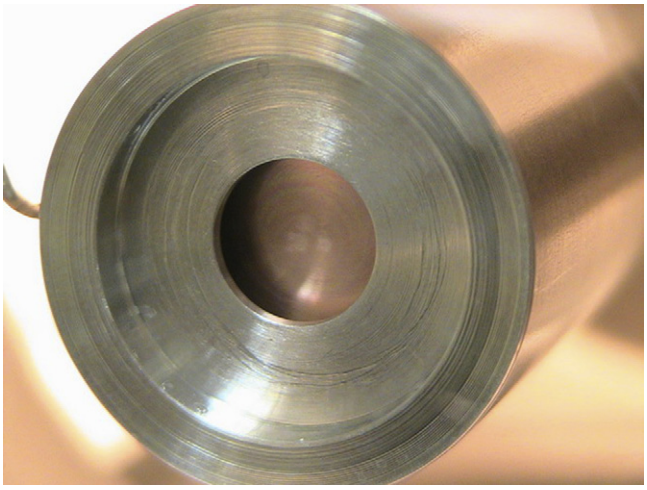

Fig. 3. Three white nitrogen spots in the center and the ring fringe pattern. The scale size is given by the 9 mm opening in the cold shield in front of the face of the Cu cold finger. The condensation spots are about 1 mm in diameter, which is close to the size of the opening in the gas deposition nozzle.

previous equation is rewritten as:

$$S = 1 - \frac{P_2 - P_0}{P_1 - P_0}$$

P_0 was given by fitting the last falling pressure curve in Fig. 1 with a three component exponential decay. For each P_1, the fitted P_0 is not the same because the evaporation is relative to the amount of nitrogen condensed on the surface. For this reason, the experiment was conducted from low to high initial pressure so that the effect of evaporation was minimized. We did not make fitted curve to find the limiting value of the pressure corresponding to P_2 because there were not enough data points for an unambiguous extrapolation. Instead we have taken as P_2 the pressure reading 1 min after moving the nozzle to its position directly in front of the cold finger surface. Given the slope of the curves in Fig. 2, this is clearly an upper limit and the S's calculated with these values of P_2 thus give lower limits for the sticking coefficient. The sticking coefficients calculated in this way are shown in Fig. 2.

The plot of S versus P_1 (Fig. 2) indicates that the smaller values of S at low values of P_1 are probably erroneous due to P_2 and P_0 both being close to the base pressure of the vacuum chamber which is easily perturbed by the admission of various amounts of nitrogen gas. The unweighted mean sticking coefficient, excluding the two values obtained at the lowest readings of P_1, yields an average $S = 99.0 \pm 0.6\%$, where the error estimate is the standard deviation of the list of five measurements. Systematic errors from finite slopes at the points B, as in Fig. 1, suggest that S could be in fact as much as 0.5% higher.

When the nitrogen pressure was as low as 3.2×10^{-8} Torr, a small nitrogen spot could be observed by eye following the gas exposure, indicating the presence of a thick layer of nitrogen. At higher pressures, a white nitrogen spot could be seen and stayed unchanged for much longer than the 10 min, which would correspond to the half-life of ^{13}N. In Fig. 3, three such spots are seen along with a colorful interference fringe pattern, which was caused by the spreading of nitrogen molecules to adjacent areas to form a thin film.

4. Conclusions

At room temperature, the sticking coefficient of nitrogen on an uncharacterized copper surface is very small because only one or possibly two monolayers of nitrogen can be adsorbed on the surface. At temperatures of order 12 K, macroscopic quantities of nitrogen condenses forming a visible nitrogen spot, which persists for much longer than the lifetime of ^{13}N. The spot appeared white, indicating that it was thick and polycrystalline. The average sticking coefficient of nitrogen on the solid nitrogen film is close to unity ($99.0 \pm 0.6\%$).

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