approximation³ to the Hill and Van Vleck formula which yields the relation

$$T_1'(K) - T_2'(K) = [A\Lambda^2/K(K+1) + \gamma](K+\frac{1}{2})$$

is not expected to apply when the ratio A/B is so far from the value 0, we find that an equation of this form with A = 9.3 and $\gamma = +0.230$ quite accurately gives the doublet intervals of Fig. 1 up to K = 20. We therefore take these values as the approximately correct ones for the coupling constants of this E state.

The size of the γ is indicative of large interaction with other near-lying CaH levels, largely, perhaps, those of the *D* state whose v=0 level lies but 2135 cm⁻¹ above the origin of this *E* band. In view of the violent perturbations encountered in these terms, the changing value of γ which we note for the higher *K* levels is not surprising.

Fig. 2 is a plot of the Λ -doubling in this ²II state. In sign, relative position and magnitude these curves are about what one would expect for a ²II state of this character. These values of $\Delta v_{1dc}(J)$ and $\Delta v_{2dc}(J)$ yield the constants $p_0 = +0.052$ and $q_0 = +0.017$ upon application of the well-known Λ -doubling formulas.⁴ It is not to be expected, however, that a relation of "pure precession" should hold between this state and a single other CaH state.

The Q_1 and Q_2 branches of the (0,0) band form ³ Reference 2, p. 508.

⁴ R. S. Mulliken and A. Christy, Phys. Rev. 38, 87 (1931).





heads at 20,410.60 and 20,406.11, respectively. A similar pair of fainter heads for the (1,1) band occur at 20,337.16 and 20,333.65, while corresponding Q branch heads of the (2,2) band lie at 20,258.66 and 20,254.30. Assuming the small interval between the origin and Q head to be the same for each of these bands, one can compute the vibrational constants for this ²II state given in Table II. Just as for the D state,¹ the relation $D=4B_0^3/\omega_0^2$ which holds accurately for the normal state fails to hold for this E state. This fact indicates again the existence of strong perturbing influences from neighboring CaH levels.

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Photoelectric Properties of Pure and Gas-Contaminated Magnesium*

R. J. CASHMAN AND W. S. HUXFORD, Department of Physics, Northwestern University (Received August 19, 1935)

The present study is a continuation of an earlier investigation by the authors on the effects of gases on the photoelectric sensitivity of magnesium. A method for purifying magnesium by multiple distillations is described. The threshold characteristic of all gas-free surfaces obtained by successive distillations is found to lie at $3430\pm20A$. The threshold value at 5100A, previously ascribed to pure magnesium, is now definitely attributed to magnesium contaminated by hydrogen. The effects of oxygen are further investigated in detail, a very slight dosing giving rise to a maximum excursion of the long wave limit to about 5700A. Calculations show that the formation of polar molecules on the magnesium surface will account for the threshold shift observed when hydrogen or oxygen is present.

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 \mathbf{T} N a previous report by the authors¹ on the photoelectric properties of magnesium it has been shown that layers of this metal deposited in vacuum exhibit rapid changes in sensitivity when slight traces of air or oxygen come in contact with the surface. Pure nitrogen had no effect on the emission except when a glow discharge was set up in the gas, so that the sensitization caused by air was attributed solely to the oxygen contained in it. These results appeared to account rather well for the large discrepancies between thresholds for magnesium reported by many investigators in the past 15 or 20 years, but left unconfirmed recent values obtained for metal supposedly gas-free and in the pure state. The investigation was continued in the hope that with new refinements in the technique of purification and degassing of the metal completely gas-free layers could be obtained. This anticipation has been achieved with respect to the elimination of traces of gas which affect the photoelectric sensitivity of magnesium. In addition the investigation includes a detailed study of the surface structure of the metal when activated by contact with oxygen or hydrogen.

Gas-Free Surfaces of Magnesium Formed by Multiple Distillation

An extended study of magnesium surfaces prepared by single distillations of the metal indicated the presence of traces of residual gases too small to be detected by pressure-measuring devices. A search was undertaken to discover what gases are easily absorbed and probably, therefore, easily retained by the condensed layers. It was found that heated magnesium absorbed large volumes of hydrogen upon cooling. This observation was not in agreement with the work of Mrs. M. R. Andrews² but was in accord with observations of Hanawalt,3 who found that large volumes of hydrogen were evolved by ordinary specimens of magnesium when they were heated. A more recent publication by Reimann⁴ gives a detailed account of the "clean-up" of hydrogen by magnesium.

It is our experience that a condensed surface formed by rapid sublimation of magnesium is always contaminated with gas. A study of the photo-emission from layers deposited by slowly subliming the metal in successive distillations showed that the gas which is evolved during this time is hydrogen, although the sample had not been previously exposed to the gas. A subsequent slight contamination of such surfaces with hydrogen results in a definite and reproducible shift of the threshold, indicating a sensitization arising solely from the action of this gas upon the pure metal. Moreover, the maximum excursion of the threshold so obtained agrees exactly with the threshold at 5100A previously obtained¹ for magnesium layers which had not been specially treated to remove the hydrogen.

The method of successive distillations involves slow vaporization of a considerable quantity of magnesium in an auxiliary chamber in which a portion of the metal as it condenses is collected on a gas-free molybdenum or tantalum plate. At low gas pressures appreciable dispersal gettering does not occur if the vaporization is carried out very slowly. If the collecting plate is close to the vaporizing sample, the deposit on it is thicker than the layers formed at larger distances on the walls of the glass bulb. It is obvious that these latter layers of larger area will remove by contact gettering the major portion of hydrogen gas evolved during the process.⁵ The hydrogen absorbed per gram of magnesium collected on the plate will be small by comparison to that absorbed per gram of metal condensed on the walls of the bulb. The plate is then removed to a second chamber, and the process repeated, the magnesium being again collected on a second movable plate.

An extension of the study in which condensed layers of magnesium were deposited by a single distillation showed that under certain conditions effectively gas-free layers may be formed. The magnesium specimen is first given a very slow progressive heating while the tube is still on the pumps. After several hours and at the time when noticeable sublimation has begun the tube is sealed off. A coating is then formed by very

¹ R. J. Cashman and W. S. Huxford, Phys. Rev. **43**, 811 (1933).

² Quoted by Dushman, Rev. Mod. Phys. **2**, 381 (1930). ³ Hanawalt, Dow Chemical Co. Private communication.

⁴ Reimann, Phil. Mag. 16, 673 (1933).

⁵ Reimann, reference 4, found that one hydrogen molecule out of every 1.5×10^6 which impinge upon the surface of magnesium is absorbed at room temperature.

slow sublimation. This coating is gas-contaminated but subsequent coatings formed in the same way show the emission characteristics of pure magnesium.

Experimental Photo-Cells; Outgassing Treatment

It is well known that if the temperature of an evacuated glass bulb containing magnesium is increased to a point where sublimation of the metal is noticeable (about 400°C) a reaction occurs between the glass and the magnesium, and a brown or black deposit forms along the inner wall. It is necessary because of this reaction to develop other methods of distillation so that the glass is never heated to a high temperature while in contact with magnesium.

The first type of tube used for the study of multiply distilled surfaces is shown in Fig. 1.

Since the tube contained several metal parts, extreme care had to be exercised in outgassing. Following the usual cleaning of glass and metal parts the tube was sealed to a two-stage mercury diffusion pump containing two liquid-air traps. An oven at a temperature of 550°C surrounded the tube for a period of 10 days. During this time the long molybdenum tube D was heated to 2000°C many times with an induction furnace, and the other metal parts were subjected to a vigorous heat treatment by electron bombardment.

At the end of 10 days dried tank-nitrogen was allowed to fill the system and the vertical side tube A was broken off for the purpose of admitting the magnesium. Through the courtesy of the Dow Chemical Company, magnesium was secured which had been especially purified by fractional distillation in an evacuated iron tube. The specimens were better than 99.9 percent pure and contained traces of iron and aluminum only. About 3 grams of the metal were introduced into the inner Pyrex tube C.

Another week of outgassing followed. An auxiliary oven maintained at a temperature of 340° C was mounted around the side arm containing the magnesium. The remainder of the tube was kept at 550°C as before. During the final stages the metal parts were given a thorough heat treatment. Magnesium was finally introduced into the vertical tube *D* by moving the



FIG. 1. Diagram of tube used for study of multiply distilled surfaces of magnesium. A, side tube for admitting magnesium; B, iron armature sealed in Pyrex tube; C, movable tube containing magnesium; D, molybdenum tube 35 cm long, 7 mm diameter; E, molybdenum plate 1.8 cm $\times 1 \text{ cm} \times 5 \text{ mil}$; F, 300 cm³ flask containing 13-mil spiral tungsten filament; G, molybdenum plate, 1.8 cm $\times 1 \text{ cm}$ $\times 5 \text{ mil}$; H, 300 cm³ flask containing 13-mil tungsten filament; I, thin Pyrex window 0.01 mm thick; J, side tube connection to pump; K, side tube for admitting gas, tip broken by an iron armature enclosed in glass.

tube C into position by electromagnetic control. A slight tapping of the tube caused the pieces to fall into the vertical tube.

The magnesium was slowly heated in successive stages with the induction furnace. The coil of the furnace was slowly raised and the magnesium repeatedly distilled and condensed until a mirror-like coating formed at the top of the outer glass tube. The chamber was sealed off from the vacuum system at this point, following which the molybdenum plates were heated to the point where vaporization of the molybdenum began. It was believed that any residual gas would be effectively removed by the "gettering" action of the condensed magnesium and molybdenum.

Magnesium was again vaporized from the bottom of tube D and allowed to condense on plate E. This plate was moved to bulb F where a very gradual heating of the filament caused some of the magnesium to sublime and condense on plate G. Sensitivity plots were obtained both for coatings formed in this manner and for coatings formed on the wall of bulb H.



FIG. 2. Diagram of second type tube used for study of multiply distilled surfaces of magnesium. (a) A, highly insulated anode; B, nickel leads; C, contact to cathode; D, tungsten wire, makes contact with plate during electron bombardment; E, tungsten wire seals; F, 13-mil spiral tungsten filament 1 cm² area; G, Pyrex shields; H, tube for admitting gas; I, contact to cathode; J, 13-mil coneshaped spiral tungsten filament for vaporizing magnesium; K, nickel plates to prevent magnesium from going down neck; L, tantalum plate 1.8 cm×1 cm×5 mil; M, Pyrex rod; N, iron armature. (b) O, thin Pyrex window (one on each bulb perpendicular to (a); P, Pyrex shield.

Another tube possessing the advantages of a minimum number of metal parts and high electrical insulation is shown in Fig. 2. Only two distillations were possible but each deposit could be studied individually. A third tube contained simply a tungsten spiral filament inside of which the magnesium sample was placed, and layers were deposited on the glass walls in a single slow distillation.

MEASURING EQUIPMENT

The photoemission was measured with an FP 54 Pliotron amplifier. With a Leeds and Northrup high sensitivity galvanometer in the plate circuit, currents as small as 2×10^{-16} ampere were measurable.

Light was resolved with a Gaertner quartz monochromator, a 68-watt linear spiral tungsten filament ordinarily serving as the light source. Qualitative observations were made in the range 2000 to 2800A with iron and mercury arcs.

RESULTS

Work function of pure magnesium

The composite curve of Fig. 3 shows typical total emission plots obtained with the three tubes. Ordinates have not been reduced to unit light intensity, but have been adjusted so that the maximum current is the same for all tests.

Fig. 4 illustrates how small traces of gas remaining in a tube following the sealing off process may affect the emission from the first deposit of magnesium. All later deposits in this bulb, and all layers formed in the other two types of cells, both for magnesium surfaces formed on molybdenum or tantalum plates and on the walls of the Pyrex bulbs, are in excellent agreement and fix the threshold at 3430A, approximately, corresponding to a work function of 3.60 volts.

When illuminated with resolved light from a tungsten filament the emission from these pure magnesium surfaces was so small that the total available sensitivity of the amplifying circuit was required to obtain readable deflections. Under these circumstances it was not possible to obtain the energy distribution of the resolved light and apply Fowler's method for determining the true work function. A consideration of the finite range of wave-lengths passing the slit of the monochromator leads to an estimated error of $\pm 20A$ in the apparent threshold determinations.

Work functions for tantalum and molybdenum

An excellent opportunity for measuring the photoelectric work functions of tantalum and



FIG. 3. Composite curve showing results from three cells.



FIG. 4. Curves showing the "clean-up" of residual gases in a tube following the sealing off process.

molybdenum was provided by the multiple distillation tubes. By using the full sensitivity of the amplifier and taking the last observable reading as the approximate value of the threshold, a value of 3000A (4.12 volts) was obtained for molybdenum and 3050A (4.05 volts) for tantalum. DuBridge and Roehr⁶ obtained 4.15 volts for molybdenum and Cardwell⁷ obtained 4.10 volts for tantalum.

Hydrogen on magnesium

Samples of hydrogen were obtained by heating specimens of ordinary magnesium in vacuum and from an outside tank supply. In the latter case the hydrogen was allowed to flow through a calcium chloride drying tube, a liquid-air trap, a small capillary tube onto a glowing platinum spiral and through another liquid-air trap before introduction into the photo-cell.

Results when using either source for the gas showed an immediate increase in sensitivity and a shift of the threshold from 3430 to 5100A. Pressures of hydrogen ranging from about 10^{-6} or lower to 10^{-1} mm of Hg produced the same shift in the threshold. Fig. 5 shows the effect of admitting different amounts into the cell. The first trace of gas gave rise to a selective maximum at about 4100A and a threshold of 5100A. Pumping practically removed the maximum and produced



FIG. 5. Effect of hydrogen on magnesium.

a shift in the threshold to about 4700A. An added dose, however, gave the characteristic value at 5100A, but the maximum either disappeared or was masked by the large increase in sensitivity over the range 2800–5100A. Pumping again caused the threshold to recede to 4700A.

The above phenomenon was observed several times, the presence of a trace of hydrogen in the cell giving rise to a threshold at 5100A whereas the absence of the trace gives the value 4700A. Finally a small amount of gas was admitted and caused to dissociate by means of a glow discharge. The threshold again receded from 5100 to 4700A and a rapid "clean-up" of the gas took place. When the magnesium was distilled in the *presence* of hydrogen a characteristic threshold at 5100A always appeared.

These results indicate that at least two definite equilibrium conditions may exist at the surface under the action of hydrogen, one of which is characterized by a threshold at 4700A and is probably due to the effect of atomic hydrogen alone, and the other of which gives rise to a threshold at 5100A and is probably due to both atomic and molecular hydrogen. These results are discussed in more detail later.

⁶ DuBridge and Roehr, Phys. Rev. 42, 52 (1932).

⁷ Cardwell, Phys. Rev. 38, 2041 (1931); 47, 628 (1935).

It was possible to obtain the energy distribution of the resolved light in the region near 5100A and consequently Fowler's theory could be applied to an activated surface at room temperature. The experimental points were found to fit the theoretical curve and the threshold was shifted from the apparent value 5100A to the true value at 5040A. Others⁸ have obtained similar checks on the theory using activated surfaces so that it seems established that the theory may now be applied to homogeneous activated surfaces as well as clean surfaces.

Oxygen on magnesium

Considerable difficulty was encountered in obtaining pure oxygen. Spectroscopic examination of samples prepared in various ways generally indicated the presence of minute traces of hydrogen and carbon monoxide. A method was finally developed by which oxygen was obtained from mercuric oxide by heating.

The oxygen passed through a liquid-air trap to a jet, from which it streamed against a glowing platinum spiral and through another liquid-air trap to the photo-cell. A spectroscopic analysis with exposure times as long as 15 hours revealed only the presence of oxygen. At room temperatures the mercuric oxide evolved no oxygen so that a controllable heater provided a convenient means of obtaining very small traces of the gas.

The sensitization and final desensitization of a magnesium surface by oxygen is shown in the curves of Fig. 6. The initial sensitization occurred at a pressure of 10^{-7} mm of Hg or less. The maximum shift in the threshold toward the red was from 3430 to about 5700A (see curve III). Further exposure produced a decrease in sensitivity and a recession of the threshold toward the violet. Pumping, however, brought the threshold back to 5700A. As long as the gas pressure was small it was found that this phenomenon could be repeated.

Finally oxygen was allowed to flow slowly into the cell. As the threshold receded into the ultraviolet, curves IV, V, VI and VII were taken. The procedure was to set the monochromator at different wave-lengths as quickly as possible and determine the corresponding emission currents. Since the sensitivity was continually changing the curves are only rough approximations. Eventually the threshold receded to a value below 2000A and pumping the tube failed to restore the sensitivity.

Oxygen and hydrogen on magnesium

If a layer of magnesium is laid down in the presence of slight traces of hydrogen and oxygen in the tube, remarkable increases in sensitivity and decreases in work function may be observed. The response to unresolved light may be as high as 10^6 times the response of the pure layer, and the corresponding threshold has been found as far out as 7000A. The *yield* to white light, however, is very small when compared with ordinary commercial caesium oxide cells.

DISCUSSION OF RESULTS

These experiments show that the threshold 5100A obtained previously by the authors¹ is characteristic not of pure magnesium but of magnesium contaminated with hydrogen. In the early work sufficient time was not spent during the heating and vaporizing period. It is now obvious that when hydrogen and magnesium come off together from a specimen of the metal at a rapid rate, dispersal gettering occurs and a condensed layer containing both elements will be formed.



FIG. 6. Effect of oxygen on magnesium.

⁸ DuBridge, Phys. Rev. **46**, 339A (1934) (sodium on nickel); Jamison, unpublished results (thoriated tungsten).

On the other hand, the constancy and reproducibility of the threshold 3430A, obtained from surfaces formed by repeated distillation and very slow sublimation following the methods given above, provide convincing evidence that the layers are gas-free. The result is in good agreement with the values 3400A obtained by Déjardin and Schwegler⁹ and 3300A, obtained by Kenty,¹⁰ for pure magnesium surfaces.

That the discordant results obtained by other investigators arise from contamination by hydrogen or oxygen, or both, is shown by the gas study. All the published values fall within the limits set by the present work, which range from 2000A or less for heavily oxidized magnesium to 7000A, the threshold observed when magnesium is distilled in the presence of traces of hydrogen and oxygen. The high photoelectric efficiency for magnesium reported by de Laszlo¹¹ (1 electron per 100 quanta at 3130A) as compared to the low value found by Déjardin and Schwegler⁹ (1 electron per 25,000 quanta at 3130A) may be attributed to activation by hydrogen or oxygen, since the sensitivity at 3130A has been increased several hundred-fold by the authors through application of traces of these gases.

Effect of oxygen on other metals

Oxygen has the effect of increasing the work function of electronegative metals, the most noted example being that of tungsten.¹² Others¹³ have obtained similar results for gold, silver, palladium, platinum, tantalum and molybdenum. For the more electropositive metals, however, most observers report a sensitization by small traces of oxygen. Olpin¹⁴ and Selenyi¹⁵ have produced shifts in the threshold of sodium of about 1000A toward the red. Koller¹⁶ and others have sensitized caesium with oxygen. Renschler and Henry¹⁷ have lowered the work

¹⁰ Kenty, Phys. Rev. 44, 891 (1933).
¹¹ de Laszlo, Phil. Mag. 13, 1171 (1932).
¹² Langmuir, J. Am. Chem. Soc. 38, 2221 (1916);
¹³ Hughes and DuBridge, *Photoelectric Phenomena* (McGraw-Hill, 1932), p. 78.
¹⁴ Olpin, Phys. Rev. 36, 251 (1930).
¹⁵ Selenyi, *Photoelectric Cells and Their Application*

functions of uranium, calcium and thorium by exposing these elements to traces of oxygen whereas heavy dosing completely destroyed all photoelectric activity.

Interpretation of results for magnesium

Calculations based on the Sommerfeld model for the electrons in a metal show that changes in the inner work function, W_i , under the action of absorbed gases, are too small to account for the large changes in work function observed. It is believed that the phenomena are due to changes in the character of the surface of the metal, presumably by the formation of an electric double layer and hence the observed results are attributed to changes in the outer work function, W_a .

To account on a classical basis for the observed decrease in work function when a sparse distribution of atoms of an alkali or alkali earth metal are deposited on an electronegative metal base such as tungsten, Langmuir, Becker, Suhrmann, De Boer and others have assumed an electric double layer, formed by + ions of the electropositive material and their images in the metal underneath. If the ionization potential of the adsorbed particle is much higher than the work function of the base metal experiments have shown that the work function is increased.

Thus in our experiments an adsorbed hydrogen molecule (ionization potential, 16 volts) or an adsorbed oxygen molecule (ionization potential, 12.5 volts) certainly would not be expected to give up an electron to the magnesium undercoating (work function, 3.60 volts). If dissociation of the molecules takes place the ionization potentials of the resulting atoms are still too high (13.6 volts for O and 13.5 volts for H). Even if one assumes that magnesium atoms in some way detach themselves from the metal, get on top of the adsorbed gas layer and a portion become positive ions (ionization potential 7.61 volts) the work function should vary continuously with the number of adsorbed molecules, a conclusion in agreement with the oxygen experiments, but not the hydrogen experiments.

In order to account for the observed results on a classical basis we may assume that polar molecules are formed at the surface between magnesium atoms and atoms of the gas. This

⁹ Déjardin and Schwegler, Comptes rendus 196, 1585 (1933)

¹⁰ Kenty, Phys. Rev. 44, 891 (1933)

⁽Sir Isaac Pitman and Sons, 1929), p. 25.

¹⁶ Koller, Phys. Rev. **36**, 1639 (1930).

¹⁷ Renschler and Henry, Phys. Rev. 47, 807A (1935).

assumption seems reasonable since O_2 or H_2 possess no permanent dipoles and under the influence of van der Waals forces alone at the surface would not have any dipoles induced in them. On the other hand nonhomonuclear molecules as a rule possess permanent dipoles. By making assumptions as to the orientation and number per cm², the value of the dipole for the different molecules is calculated below, and the results indicate the correct order of magnitude when compared to similar molecules.

For hydrogen on magnesium the threshold shift from 3430 to 4700A is attributed to the formation of a single layer of MgH molecules on the surface having a positive component of the dipole outward from the metal and the number per cm² the same as the number of Mg atoms per cm² at the surface. The experiments indicate that this surface layer of molecules is permanent and any hydrogen which diffuses into the metal must come from the outside. The second shift from 4700 to 5100A obtained when traces of hydrogen are admitted is attributed to induced dipoles in weakly adsorbed hydrogen molecules. An equilibrium condition must then exist at the surface between molecules coming in from the outside, molecules leaving the surface outwardly and molecules or atoms diffusing into the interior.

The change in potential experienced by an electron in going through an electric double layer is $\Delta \varphi = 4\pi NP$, where N is the number of dipoles per cm² and P the dipole moment. By taking 1.59A as the radius of the magnesium atom (from crystal structure data) the number of MgH molecules per cm² is assumed to be

$$N_{\rm MgH} = (1.59 \times 2 \times 10^{-8})^{-2}$$
 per cm².

If 4700A (2.62 volts) is the threshold corresponding to this maximum surface coverage, the value for $\Delta \varphi$ is 3.60-2.62=0.98 volt or 3.3×10^{-3} e.s.u. Assuming that all of the normal component of the dipole contributes to the change in work function,

$$P_{\rm MgH} = \frac{\Delta \varphi}{4\pi N} = \frac{3.3 \times 10^{-3}}{4\pi (1.59 \times 2 \times 10^{-8})^{-2}} = 0.26 \times 10^{-18} \text{ e.s.u.}$$

The maximum shift in the threshold of magnesium from 3430 to 5700A when exposed to oxygen is assumed to be due to the formation of a single layer of MgO molecules oriented with a positive component of the dipole outward from the metal. The final desensitization in which the threshold recedes to a value below 2000A is attributed to a thick coating of MgO, the emitting layer being characteristic of this compound and not of magnesium.

If it is assumed that the number of MgO molecules per cm² is the same as the number of magnesium atoms per cm² of surface, similar assumptions and calculations as were made above lead to the value

$$P_{\rm MgO} = 0.38 \times 10^{-18} \text{ e.s.u.}$$

In like manner the shift of the threshold of magnesium from 3430 to 7000A when exposed to traces of oxygen and hydrogen is assumed to be due to the formation of a single layer of MgOH molecules. The value of the dipole moment of this molecule, using similar assumptions, becomes

$$P_{\rm MgOH} = 0.51 \times 10^{-18} \text{ e.s.u.}$$

The magnitudes of these dipole moments fall within the range of measured values of the moments of several nonhomonuclear molecules for which experimental data exist. If there is appreciable deformation of the molecules adsorbed on the surface their dipole moments may differ somewhat from the measured values for the same molecules in the gas state. The agreement in order of magnitude of the moments calculated from the change in photoelectric threshold and those of similar molecules measured by other methods, provides considerable evidence for the existence of nonhomonuclear molecules on the surface of magnesium contaminated with certain gases.