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# Recent Results in Particle Adhesion: UHV Measurements, Light-Modulated Adhesion and the Effect of Adsorbates<sup>†</sup>

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In the past few years, the force of adhesion  $F$  between small metal spheres (about 2 to 8  $\mu\text{m}$  in diameter) and flat semiconducting or metallic substrates has been measured under a variety of conditions. Most of this work has been or is about to be published. This is a summarizing review of the results obtained. An ultracentrifuge technique has been employed. In measurements under ultra-high vacuum the adhesion between Au spheres and flat Si substrates was studied as a function of the oxide layer thickness on Si. Also flat Au substrates were used. The results fit with the Lifshitz theory of van der Waals forces. In another series of measurements the van der Waals component  $F_{vdW}$  of  $F$  was separated from the electrostatic one  $F_{el}$  which originates from the electrostatic double layer formed at the interface of the adherents. This was achieved by varying  $F_{el}$  by band-gap light of varying intensity. The adherents were Zr-coated Au spheres on CdS.

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## MEASURING TECHNIQUE<sup>6</sup>

Substrate and spheres are fixed to the periphery of a rotor of an ultracentrifuge capable of a maximum speed at which an acceleration of about  $10^6$  g is

<sup>†</sup> This paper was presented at the *Symposium on Recent Advances in Adhesion* during the 162nd National American Chemical Society Meeting, September, 1971.

imparted to the spheres. The number of spheres removed after application of a given acceleration is determined by microscopical counts at standstill of the rotor. The figure obtained is multiplied by the mass of the spheres and the resulting force of adhesion is plotted versus the percentage of particles removed.

Some measurements were carried out in ultrahigh vacuum, the others at about  $10^{-4}$  mm Hg. Room temperature was employed.

### ULTRAHIGH-VACUUM MEASUREMENTS

The results of the ultrahigh vacuum measurements are summarized in the subsequent table. In column III the median adhesive force  $F$  over sphere radius  $R$  is indicated because, according to theory,<sup>6</sup> proportionality  $F \propto R$  is expected, see below.

Substrate	Sphere diameter $2R$ ( $\mu\text{m}$ )	Median value $F/R$ (dyn/cm)	Effective Separation $z_0$ ( $\text{\AA}$ )
freshly cleaved Si (111)	2.6 ... 5.8	$172 \pm 34$	$5.7 \pm 0.6$
Si (111) + oxygen monolayer	2.6 ... 5.8	$89 \pm 10$	$7.9 \pm 0.5$
Si (111) + 40- $\text{\AA}$ oxide layer	2.6 ... 5.8	$48 \pm 2$	—
Si (111) + 130- $\text{\AA}$ oxide layer	2.6 ... 5.8	$17 \pm 7$	—
3 types A, B, C of flat Au substrates of increasing smoothness			
A	1.75 ... 5.0	$20 \pm 5$	19
B	1.75 ... 5.0	$113 \pm 30$	8
C	2.0 ... 8.0	$160 \dots 445$	4

According to theory,<sup>6</sup> the van der Waals component  $F_{vdW}$  of the adhesive force  $F$  is given by

$$F_{vdW}^* = \frac{\hbar\bar{\omega}}{8\pi z_0^2} R$$

where  $\hbar\bar{\omega}$  Lifshitz-van der Waals constant

$z_0$  effective separation sphere/substrate.

Column IV indicates the effective separations  $z_0$  between the adherents, calculated from the experimental values  $F/R$  and from theoretical values for  $\hbar\bar{\omega}$  ( $\hbar\bar{\omega}$  for Au/Si 8.67<sup>7</sup>; for Au/Au 11.86<sup>7</sup>). The value in the first row,  $z_0 = 5.7 \text{ \AA}$ , points to residual surface asperities.

The increase to 7.9 Å may be interpreted as follows. The oxygen monolayer contributes little to adhesion and acts as a spacer only. The difference between 7.9 and 5.7 Å is consistent with the ionic diameter of  $O^{2-}$  of 2.6 Å.

With gold spheres on gold substrates it was only with the smoothest substrate, C, and in particular with the largest spheres, 8 μm in diameter, that  $z_0$  was found to approach 4 Å, i.e. interatomic distances, probably owing to small surface deformations under the action of the van der Waals forces.

An extension of the *Lifshitz theory* of van der Waals forces to geometries other than plane-parallel and to *adherents covered by adsorbed films* was presented by Langbein.<sup>8</sup> A quantitative application of the formula obtained is not yet possible because of the lack of experimental data on the dielectric constant of oxide films on Si.

#### LIGHT-MODULATED ADHESION<sup>2-4</sup>

Whereas in the above systems Au/Si(O) and Au/Au electrostatic forces probably do not play a significant role,<sup>5</sup> they were shown to dominate over the van der Waals forces in the system Zr-coated Au spheres placed on low-conductivity CdS.

The experimental evidence was obtained as follows. The adhesion measurements were carried out in the dark and at different intensities of light of a wavelength of 522 nm. The illumination increased the dark conductivity by a factor of about  $10^5$ , and the median value of  $F/R$  varied by a factor of about 2. This is attributed to the light changing the quasi-Fermi level in CdS and hence the electrostatic double layer at the junction Zr/CdS. The theory yields a quadratic relationship between the electrostatic component  $F_{el}$  of the total force  $F$  and the distance  $E_n$  between the Fermi level and the conduction band of CdS:

$$F_{el} \propto [E_m - (E_a + E_n)]^2$$

$E_m$  work function of Zr

$E_a$  electron affinity of CdS

The relationship between the illumination-dependent free-carrier density  $n$  and  $E_n$  is

$$n \propto \exp - \frac{E_n}{kT}$$

In this manner, the photoconductivity and the adhesion  $F_{el}$  can be correlated. In the system Zr-coated Au spheres,  $F_{el}$  exceeds  $F_{vdw}$  by a factor of up to 2.

## CONCLUSIONS

(1) An unambiguous separation of the van der Waals contribution  $F_{vdw}$  and the electrostatic contribution  $F_{el}$  to the total force of adhesion  $F$  of a small sphere adhering to a flat substrate has been achieved experimentally. Depending on the materials used,  $F_{el}$  may be small or large compared with  $F_{vdw}$ .

(2) Although the experimental systems used were of a much better defined geometry than most adhesion systems studied so far,<sup>6</sup> a quantitative comparison of the van der Waals theory of adhesion with experiment is still impeded by small surface irregularities (cleavage steps, asperities) of the substrates. No independent measurement of the effective distance  $z_0$  between the adherents has yet been made.

(3) The theory of the effect of surface films on adhesion has not yet been tested quantitatively, since the dielectric constant of such films is not well defined and experimental data are not available.

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