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A PROPOSED NON-INTRUSIVE METHOD FOR FINDING COEFFICIENTS OF  
SLIP AND MOLECULAR REFLECTIVITY IN MICROGRAVITY

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## I. Introduction

When gas flows past solid or liquid surfaces, interesting effects occur. Figure I, Appendix A shows a typical velocity profile for a gas moving past a surface. These effects may all be included under the general heading of **slip** or **jump** phenomena. The latter term highlights the principal problem confounding their detailed experimental or theoretical explanation. Namely those properties measured in the layer connecting solid or liquid surfaces to a gas may jump or vary discontinuously. Discontinuous changes are notoriously difficult to describe. Most often in practice, bulk values are extended linearly to zero at the boundary. Here infinite stresses are ignored or neglected. No complete theory presently addresses these singularities. But in practice, average properties expected for gas moving near a surface-- properties like velocity, concentration, and temperature--will differ from corresponding properties measured at the surface. There is no reason to expect this difference to be linear. For example, the mass-averaged velocity  $v$  of gas molecules moving tangentially to a surface may not equal zero. Depending on how gas molecules reflect at the surface, velocity profiles will vary in complicated, often non-linear ways.

Kinetic theory relies on the assumption that macroscopic properties such as pressure and temperature arise from molecular forces. These forces are exerted by molecules leaving a surface. Their peculiar character depends on exactly *how* the molecules leave. Dating back a century, the work of J.C. Maxwell [1] isolated three types of surface-gas effects: 1) viscous slip; 2) diffusive slip; and 3) thermal slip. These remain hydrodynamically valid today and each requires precise determination before proceeding with standard calculations prescribed by kinetic theory.

For example, to treat each type of gas-surface effect, the usual no-slip boundary condition (stating that the velocity of a gas bounded by a surface must equal the surface velocity at every point) requires amendment [1-11]. This follows namely

when there exists a velocity gradient directed perpendicular to the surface (in the case of viscous slipping) [1,5], a concentration gradient directed parallel to the surface (in the case of diffusive slipping in a mixture having different molecular weights) [2-11], or a temperature gradient directed parallel to the surface (in the case of thermal slipping) [5,7].

All these cases follow from the assumption defining static conditions, namely that the net number of molecules flowing through a cross-section will equal zero. For a one-component gas, this assumption implies zero molar-averaged velocity measured at the surface and consequently no slip [2]. By definition it implies a similar conclusion for a binary gas made up from components having different molecular weights. However in the latter case, the assumption of no net molecular flow implies nothing about component molar velocities, non-zero mass-averaged velocity or finite momentum transfer [1]. In fact corrections to previous work [2, 9, 12, 13] show that for binary gases, both simultaneously vanishing mass- and molar-averaged velocities must contradict each other if molecular weights differ. It is this result that has generated considerable confusion in the literature. As proposed in microgravity, the drop method should quantify the magnitude of these previously neglected or misunderstood effects.

Previous ground results [1,9,14] suggest that these finite velocities found near surfaces may prove substantial for typical experimental conditions. For example, diffusive slip velocities increase sharply both for disparate molecular weights and for Knudsen numbers greater than  $10^{-2}$  [9]. One of the objectives of the proposed research is to quantify experimental conditions in which near-surface velocities can be neglected.

Each type of slip arises from its own source, whether following from pressure, concentration or temperature differences. Each accounts for molecular movement of gases travelling both in the transition layer and along surfaces. For example, viscous

slip follows from the pressure dependence of viscosity. Applying the assumptions of kinetic theory, Maxwell [1] deduced originally that air viscosity should vary independently of pressure; however surprisingly he found that viscosity fell off experimentally at low pressures. Subsequently he introduced the slip coefficient to monitor the ratio between internal and external friction--a ratio that tends to zero at higher pressures or no slip. In simplest terms, a gas tends to get a better grip on a surface as pressure increases. This viscous slipping varies proportionally to the near-surface velocity gradient. Its proportionality constant: 1) carries the dimension of length; 2) equals roughly the mean free path of the gas; and 3) like the latter, varies in inverse proportion to pressure.

Diffusive slipping, on the other hand, follows from (and is proportional to) the gradient of the relative concentration found in a binary gas mixture. Thus an inhomogeneous mixture of gases will set a suspended body in motion. The body will travel along the diffusing direction of the heavier gas, an effect both which is analogous to the better-known radiometric effect and also which increases as the mass difference taken between the two diffusing components increases. Thus motion occurring because of concentration gradients--always present in diffusing gases--invalidates the traditional no-slip condition [3].

Finally, thermal slipping follows from (and is proportional to) the temperature gradient imposed on a gas. It arises in gases not fixed in thermal equilibrium and moves a suspended body from hot to cold. Because of convection currents, ground research has quantified this radiometric effect only to an order of magnitude [15].

These types of surface effects prove critical to a host of problems. The dynamics of scattering often fix and control essential physics, whether the process involves adsorption, heterogeneous catalysis, chemical reactions, nucleation or condensation. To prove valid for atmospheric pressures and viscous gas conditions, kinetic theory and molecular dynamics depend on a precise statement defining how molecules reflect

at surfaces [1]. Reflection can take one of two simple forms [16]: either specular (mirror) reflection which conserves both the tangential wave vector and total magnitude of momenta, or diffusive reflection in which molecules are absorbed on a surface and then emitted with a Maxwellian distribution corresponding to surface temperature  $T_o$ . Figure II, Appendix A schematically illustrates this difference.

Each type of reflection supplies different slip and velocity results. For example, total diffusive reflection gives maximum surface friction and therefore minimum slip [17]. More specifically accurate determination of slip would furnish the complete justification for Maxwell's introduction [1] into kinetic gas theory of the coefficient  $f$ , or fraction of total molecules specularly reflected. Such a statement garnered from current molecular beam studies [16] proves valid only for near-vacuum conditions.

A familiar experiment to test these slip phenomena immerses solid or liquid surfaces in a gas. The arrangement looks at consequences expected for encounters between gas molecules colliding against surfaces. Figure III, Appendix A schematically illustrates the slip coefficient. From observing the effects resulting from gas encounters, microscopic properties such as slip distance  $\zeta$  and type of molecular reflection can be inferred. Of experimental interest here, these collisions particularly control steady state macroscopic properties such as the terminal velocity  $v$  expected for falling spheres. Thus by varying bulk properties one can derive the nature of surface encounters. On earth, the small scale of these surface effects makes them unobtainable otherwise except under near-vacuum and for non-viscous gases.

The simplest and most elegant method to study these surface effects is the classic oil drop method. R.A. Millikan first used this method to show that the electric charge  $e$  varies by an integral and multiple progression. The essence of the oil drop method consists in repeatedly changing the charge  $e$  placed on a given drop. At first, the drop picks up a frictional charge, but subsequently a radium source allows the capture of ions generated in any multiple or sign. Parallel electric plates suspend this charged

drop at different velocities  $v$ . Since velocity varies proportionally to charge, both terminal velocity  $v$  and charge  $e$  should follow the same multiple progression. If Stokes' Law predicting the drag force  $F$  on a drop proves correct, the greatest number dividing this series of charges ( $e, 2e, 5e, -3e$ , etc.) should show the absolute value expected for the elementary electrical charge  $e$ . But as Millikan found, Stokes' law does not hold strictly for drops having small radii  $r$  (comparable to the mean free path  $l$  of the gas). Here it is surface effects that begin to control drop velocities  $v$ . However this result opens up the experiment as a precise way to study a fascinating array of surface problems.

Specifically the drop method proves capable of measuring jump phenomena. Table I, Appendix A shows typical data to be collected. This precisely gives the coefficient of slip  $\zeta$  expected between gases and the surface of a liquid or solid. As outlined in the following, the derivation is roundabout, but ingenious. It is found that as the drop radius  $r$  decreases (taken relative to the mean free path  $l$ ), the measured charge  $e_i$  increases. Table II, Appendix A shows typical corrections for decreasing radii. Plotting this measured charge  $e_i$  as a function of the ratio taken between the mean free path  $l$  and radius  $r$  ( $e_i$  vs.  $l/r$ ) gives a straight line. Figures IV and V, Appendix A shows typical data graphed for a variety of drop radii. With much certainty and precision, the slope of this line gives the slip coefficient  $\zeta$ . Graphically the procedure is equivalent to reducing the medium to a condition of infinite pressure, in which Stokes' original hydrodynamic equation proves rigorously valid. To date this method has proved convenient to find oil surface properties within an order of magnitude. But the need to understand surface properties both with great accuracy and for a host of useful materials (e.g., solid spheres, liquids of uncertain density, non-spherical and faceted crystals) motivates the current work.

In summary, the drop method can test explicitly hypotheses that prove critical for both kinetic theory and molecular dynamics. It can supply: 1) a strictly valid statement defining how gas molecules reflect from solid and liquid surfaces; 2) an accurate

determination of coefficients found for viscous, diffusive and thermal slip; 3) an accurate and density-free prediction of particle settling rates measured in the absence of convection; and 4) a range of typical parameters which specify appropriate operating conditions for applying the no-slip boundary condition and for assuming total specular or diffusive reflection. In these findings, the absence of gravitational acceleration serves principally to eliminate both buoyancy-driven convection drifts and the need for density measurements. Both these can introduce errors equalling an order of magnitude on ground. Previous work has matured the relevant experimental techniques, but falls short of the accurate and elegant test of kinetic theory proposed for the better defined conditions expected in space.

## II. Expected Results

### A) Ground-based

The simple form of Stokes' law relies on the assumption that there exists no slip at the bounding surface between the medium and falling drop. [18]. However, for a sphere of sufficiently small radius suspended in a viscous medium, an appreciable correction factor appears. It varies linearly with the ratio taken between the gas mean free path and sphere radius ( $l/r$ ) and proves valid up to  $l/r=.5$  [18]. When found in the absence of this slip correction term, measured terminal velocities will exceed their expected values and substantially increase the calculated charges (up to forty percent of predicted value [17]). In simple terms, the drop falls more freely as friction decreases. Consequently this gives a very sensitive method to detect the presence of slip.

An uncorrected force balance for a charged sphere (ne charged, radius  $r$ ) falling with constant velocity  $v_i$  both in the presence of a viscous medium ( $\eta$ ) and electric field  $E_i$  is:

$$v_i = n_i e E_i / 6\pi\eta r.$$



The sphere obeys a drag force proposed by Stokes, namely that the drag varies proportionally to the terminal velocity. Here the subscript  $i$  indicates that the velocity is measured for trial  $i$ . Different trials can indicate a change in charge  $e$  by ionization or in the electric field  $E$ . For known values of  $E$ , two such trials indicate the relative ratio of charges  $e_i$  added or subtracted by ionization. Using this method repeatedly gives a simple way to find which drop run carried a fundamental charge (i.e., the least common denominator). The value of this charge can then be determined within the known accuracy of air viscosity (1 part in a 1,000).

For drops of different radii, the corrected drag force [ref.] gives the following relation between the fundamental charge  $e$  and calculated charges  $e_i$ :

$$e (1 + \zeta l/r) = e_i.$$

It is worth noting that in the absence of gravity, this correction differs by a power (2/3) from its classical counterpart [17]. Here,  $\zeta$  is the slope of  $e_i$  plotted against the ratio  $l/r$ . It is the coefficient of viscous slipping. In Appendix A, figures 1 and 2 show how this determination is performed graphically.

From the calculated coefficient of slip follows a simple determination of the fraction  $f$  of molecules diffusively reflected from the surface. Maxwell [1] used kinetic theory to show that the slip coefficient equals:

$$\zeta = (2\eta/c\rho) [(2/f)-1].$$

Here,  $\eta$  is the medium viscosity,  $\rho$  is the medium density, and  $c$  is the mean molecular speed (found from temperature). This form allows an easy determination of the percentage of molecules reflected either by a specular or diffusive mechanism.

## B) Microgravity

The absence of gravity simplifies the understanding of phenomena dominated by interfacial physics. As summarized in Table III, Appendix A, there are four reasons to study surface effects using the drop method in low gravity. First, microgravity eliminates drop drifts owing to buoyancy-driven convection. If for example the time of fall taken over a distance of 2 mm is 20 minutes it requires an extraordinary degree of stagnancy to prevent a drift of say 0.2 mm due to convection. But this would introduce an error of ten percent into the measured charge  $e_i$  [17]. The small drops ( $l/r \rightarrow 1$ ) of interest here particularly prove sensitive to any slight lack of air stagnancy. Second, microgravity eliminates drop drifts owing to misalignments between the gravity  $G$  and electric field  $E$  vector. These drifts can occur in a line taken parallel to the line of sight, such that they prove hard to detect and quantify. Measuring over a single trajectory, previous work has shown up to a ten millimeter drift arising from small misalignment [17]. Third, microgravity eliminates the need to know sphere densities  $\rho$ . This simplification adds significant accuracy to the method. Uncertainties in density introduce a ten-fold multiplier into the result. For example, a variation of 0.1 percent in density would make a difference of one percent in the result [19]. In low gravity however, the drop method can quantify surface effects found both for solids and for liquids of indefinite or unknown densities, compositions and temperatures. Uncertain densities used previously for sulphur spheres have invalidated an otherwise complete set of drop data. [17]. Finally, microgravity eliminates the need to use large electric fields ( $E/G \rightarrow 1$ ) to suspend drops. Present ground research must use large  $E$  fields to counterbalance gravity  $G$ . But in low-gravity ( $<10^{-6} g$ ), this bonus allows great flexibility. The experimenter can pick optimal conditions. For example, smaller  $E$  fields give longer measured times  $t$  for the same charge  $e$  and thus absolute timing errors  $\delta t$  diminish as a percentage of the total time ( $\delta t / t$ ).

### III. Discussion

#### A) Goals and Program Description

In the transition layer linking gases to surfaces, discontinuities found in flow parameters prove hard to characterize. When resolved to less than the mean free path  $l$  of the gas, microscale measurements tend to interfere with finding the parameters themselves. Precise controls further require low operating pressures ( $p \rightarrow 0$ ) and non-viscous gas conditions ( $\eta_{\text{air}} \rightarrow 0$ ).

However, by electrostatically suspending charged solid and liquid droplets and recording precise times  $t$  for displacements ( $\Delta x$ ), these surface effects can be understood on a molecular scale. This method is adopted here. Figure VI, Appendix A shows a typical drop trajectory. When measured in this non-intrusive way, the parameters of interest-- jump or slip distances  $\zeta$  and coefficients of diffusive reflection--can be extracted. To an order of magnitude, this method has aided understanding of various liquid-gas interfaces such as oil and water. But to date no similar characterization has proved successful for solids or for liquids of uncertain densities. Likewise, no data exists currently in either ground-based research or as part of a microgravity program that, when collected with the high accuracy expected in low gravity, could settle definitely questions outstanding in kinetic theory, molecular dynamics and cosmic physics.

When used in microgravity, the drop method accomplishes the following experimental goals:

- 1) to catch upon a minute drop and to hold under observation for an indefinite length of time one single atmospheric charge or any desired number of charges numbering between one and one hundred and fifty;

2) to show that Stokes' law governing the motion of a small sphere moving through a resistant medium breaks down as the diameter of the sphere tends towards the mean free path of the medium. (By finding the exact way it breaks down, one can extract otherwise unavailable information about surface effects such as slip and molecular reflection.);

3) to observe directly the kinetic agitation of a molecule;

4) to determine exactly the value expected for the fundamental electrical charge, one which is free of questionable theoretical assumptions and is limited in accuracy only by that attainable in the measurement found for the coefficient of air viscosity (Accurate knowledge of the fundamental charge makes possible a determination of the absolute values of all atomic and molecular weights, Avogadro's number for the absolute quantity of molecules in a given weight of any substance, the kinetic energy of agitation for any molecule excited at a given temperature, and a considerable number of other important physical properties including Planck's constant.);

5) to observe directly both the radiometric effect on solids and liquids owing to a gas thermal gradient as well as the diffusive slip arising from a gas concentration gradient.

Preparing for the 1992 flight opportunity carried aboard International Microgravity Laboratory-2 (IML-2) Spacelab mission, this undertaking describes a controlled and integrated set of fluid transport experiments. The experiments will look at a series of vapor transport properties measured along solid and liquid surfaces.

The objectives proposed for this program are:

1) with accuracy otherwise unobtainable on ground, to determine the coefficient of slip measured between gases and the surface of liquids and solids;

2) for the first time using the drop method, to classify and tabulate dominant surface effects found for a variety of solids, particularly those crystallized by vapor transport;

3) to extend understanding of settling rates predicted for cosmic dust and condensed vapor falling through planetary atmospheres.

A coordinated series of instrumented and controlled experiments would evolve according to the following matrix:

|                          | <u>Normal Gravity</u> | <u>Microgravity</u> |
|--------------------------|-----------------------|---------------------|
|                          | MSFC                  | IML-2               |
| <b>Liquid Spheres</b>    |                       |                     |
| Viscous Air              | X                     |                     |
| Binary Gas               |                       |                     |
| (concentration gradient) | X                     | X                   |
| Viscous Air              |                       |                     |
| (temperature gradient)   | X                     |                     |
| <b>Solid Spheres</b>     |                       |                     |
| Viscous Air              | X                     | X                   |
| Binary Gas               |                       |                     |
| (concentration gradient) | X                     | X                   |
| Viscous Air              |                       |                     |
| (temperature gradient)   | X                     | X                   |

## 1) Ground-based

By blowing a substance in liquid phase, a commercial atomizer will disperse very minute particles (1 to 10  $\mu\text{m}$  or larger) into the test cell. Stokes' law begins to break down as the sphere radius approaches the critical radius (equal to  $r = \eta/\rho v$ , where  $\eta$  is the coefficient for air viscosity,  $\rho$  is the air density, and  $v$  is the measured terminal velocity). To ensure that the air carrying this spray remains dust-free, its circulation first will pass through appropriate micro-filters. The initial cloud or spray will contain hundreds of drops. Following a lapse of seven or eight seconds [21], the field of view will clear completely, all except for a relatively small number of drops which have the right ratio of charge to radius. An applied electric field will select and suspend this chosen drop between parallel plates. Each plate will be optically polished to within two wavelengths of Na light [17] and sized to one part in ten thousand. Each will measure 3.5 cm in diameter and will rest from four to sixteen mm apart on ebonite posts. From a storage battery, this condenser arrangement will draw a reversible potential difference equalling up to 1000 V. It will prove possible to throw off the field and consequently to short-circuit the plates by tripping a double-throw switch. This will ensure that the plates retain no charge [27] and that the particles will stay centered in the viewing field. The investigation will assume that the potential drop due to charge leakage varies as a linear function of the time elapsed during an observation, such that by interpolation the potential applied at any time will follow. A 900 V Kelvin and White electrostatic voltmeter will record voltages. With an accuracy of .1 percent, this reading will be calibrated periodically by comparing it against NBS standards. Figure VII, Appendix A shows a schematic of plate potentials.

Once suspended inside the test cell, a drop either will form a liquid sphere or solidify. If the electric field were to distort the drop's shape, it would increase the surface area and consequently add more resistance from the medium. Since ground

work [17] has found no greater resistance encountered by drops moving against either gravity or electric fields, this effect proves entirely negligible. Further any viscous distortion would appear more pronounced for faster moving drops--a variation not in any way apparent [17]. Ground work will assume that each drop forms a perfect sphere.

Each drop will pick up first a frictional charge, then subsequently a charge of any sign or multiple discharged from an ionization source (either radium [17], X-rays [20] or corona discharge [17]). In the first case, friction involved in blowing the spray will charge droplets strongly [1, p. 68]. In the latter case, the ion count found in uncharged ordinary air will number 20,000/cm.<sup>3</sup> [18]. Each ion will carry three times the necessary kinetic energy ( $5.75 \times 10^{-14}$  ergs) to overcome the ionic repulsion of, say, sixteen charges ( $1.95 \times 10^{-14}$  ergs) [17]. But since free ion mobility (not less than 10,000 cm/s) will exceed drop mobilities greatly, all stray ions will neutralize against the plates as soon as they form. This will remain true as long as the electric field remains on. Thus charge addition will occur only when the electric field is turned off. All ionization sources will form only univalent ions with a precision not exceeding less than one percent [21]. To add a particular charge sign, either negative or positive: 1) the drop will be held close to the plate having the opposite sign; 2) the ionizing agent will supply uniform ionization; and 3) the drop will receive a shower of ions thus filtered to represent the desired sign. This entire charging procedure has been demonstrated for a single drop observed over the course of five to six hours and capturing hundreds of successive ions [22].

In total the procedure beginning with atomization, plate and drop charging, followed by velocity measurement proves extraordinarily reliable. A single drop can log up to six thousand observations [23]. Each field reversal remains free from interfering transients [24]. For example, a drop of radius 5  $\mu\text{m}$  takes less than  $3 \times 10^{-3}$  s to reach within 1 part in 10,000 of its final terminal velocity. In the case of a maximum drop velocity of 1 cm/s, the image velocity on a single filmed frame

(resolved to 400 frames per second and exposed for .00067 s) is 12 cm/s. In some cases, the position of this image can be measured to better than 0.0007 cm [24], a distance which is equivalent in time to  $6 \times 10^{-5}$  s. This gives an error in timing due to measuring the filmed image equal to 1 part in 10,000.

External sources of error will carry predictably small ranges. Apparent air viscosity will remain unaffected by strong ionization [17], a finding verified over four orders of charge magnitude. Five independent methods [25] will supply the coefficient of air viscosity to at least one part in one thousand. Any role played by internal convection either will diminish for the small drops proposed for microgravity research [26] or will disappear entirely for solidified drops. Systematic deviations arising from residual gravity can be observed and quantified by crossing the electric and gravity field vectors. Finally, by taking two speed measurements on the same drop, one coming before and one following after it has caught an ion, the investigator can eliminate entirely the properties both of the drop and of the medium. Instead, he can analyze with the greatest precision a quantity which varies exclusively and proportionally to the charge captured by the drop itself. Thus two equal measurements will eliminate errors arising from evaporation, Brownian motion, etc. In short, ground work will demonstrate the drop method as a precise and accurate means to study surface effects.

## **2) Microgravity**

### **A) NASA's KC-135 Aircraft Flights**

The component parts of the International Microgravity Laboratory's (IML-2) Bubble, Drop and Particle Unit (BDPU) slide-in test cell and minor modifications will be tested for optimum experimental conditions. While flying parabolic trajectories during repetitive low-g maneuvers, NASA's KC-135 will test operating parameters expected aboard Spacelab. Each maneuver gives from twenty to thirty seconds of



low-gravity ( $10^{-2}$  g). While these short times and residual gravity prove too limited for testing the complete experiment, it will allow testing of individual components. Specifically the electric field must be calibrated to stabilize spray inertia and select finely sized drop distributions in low-gravity. These findings will provide initial specifications and benchmark standards for instructing the Payload Specialist on proper voltage settings.

### **B) International Microgravity Laboratory (IML-2)**

During IML-2 mission, vapor transport experiments will be conducted in the Bubble, Particle and Drop Unit (BDPU). The apparatus will apply electric potentials to charged liquid and solid drops suspended between two parallel plates. It will operate at ambient Spacelab temperatures and pressures. A commercially available drop atomizer and ionization source (either radium, X-ray or corona discharge) will complement the BDPU's extensive optical viewing capability. A crewmember will monitor and manipulate each half-hour experimental run.

### **3) Sample Characterization**

When used in microgravity, the oil drop method will require minimal sample characterization. First, in microgravity surface effects need no longer depend on uncertain or unknown drop properties such as density and mass. This proves critical to studying both liquid drops carrying unknown compositions, temperatures or densities and solids formed from atomized liquids. Secondly, as long as the proportionality prevails between drag force  $F$  and terminal velocities  $v$ , drops need no longer form perfect spheres. This proves a useful simplification, particularly for solids formed quickly from atomized liquid sprays, for faceted crystals, and for low density liquids distorted by the strong plate potentials. At present, no other accurate method exists other than microgravity research to evaluate these phenomena at atmospheric pressures and viscous gas conditions.

In microgravity, the only significant requirements placed on the samples are the following: 1) that the drag force  $F$  vary proportionally to the terminal velocity  $v$ ; and 2) that the correction for small radii  $r$  taken relative to mean free gas path  $l$  vary linearly (found valid up to  $l/r=.5$  [17]). No exception currently exists in the literature, either for sample or bulk gases, that violates these simple assumptions. This is one of the biggest advantages of the drop method: in contrast to molecular beam studies, it needs no special chemical or microanalytical equipment or techniques. Further, it places no strict requirements on operating conditions such as sample purity or near vacuum. Thus microgravity opens up the drop method to study accurately and simply a host of new materials.

The most significant uncertainties expected in the experiment arise from measuring air viscosity and drop radius. Using five different methods (i.e., capillary tubes, concentric cylinders, constant deflection, pendulum, and pipe flow), ground research has found air viscosity accurately to within one part in a thousand ( $\eta=.00018240$ ) [17]. Part of the proposed ground work will verify and add further accuracy to this number. Drop radius, on the other hand, can be found by three independent means: 1) optical measurement; 2) Brownian motion [17]; and 3) calculation based both on the known elementary charge found by X-ray methods [16] and on applying an uncorrected Stokes' law ( $r=eE/6\pi\eta v$ ) iteratively.

A variety of interesting materials can both carry a frictional charge and pick up free, additional charges from ionized air. Previous work has shown the efficacy of this technique for non-conductors such as oil [17], wax [28], and shellac in alcohol solution [27]; semi-conductors such as glycerin [19]; and metallic conductors such as mercury [17, 19], platinum [19], iron [19], and Rose metal [19].

To ionize chamber air, the apparatus will include a closed tube of radium containing 500 mg. of radium bromide (activity 3,000), corona discharge or other

commercially available ionizer.

## **B) Work**

Each experiment will show that a gas may slide over surfaces with a finite relative velocity. The effect of this sliding must be to diminish the action of all tangential stresses on the surface, to leave unaffected all normal stresses, and in the course of time to set up currents sweeping over the surface [1]. These sweeping currents not only completely destroy first order solutions which arrive at simple linear or parabolic velocity profiles, but also may alter significantly the way future work looks at mass and heat transport along surfaces. For example, it is posited here that diffusion-driven currents can arise independently of bouyancy-driven convection and affect crystal growth morphology--most dramatically expected in the quiescent microgravity environment where gravity effects no longer suppress or mask these more subtle heating and diffusion effects. The latter hypothesis motivates much of the current work.

The investigators propose to conduct the following types of experiments:

### **(1) Falling sphere behavior in viscous air**

Measured for a variety of sphere radii  $r$ , total charges  $ne$ , plate potentials  $V$  and pressures  $p$ , samples of a given material, either liquid or solid, will be atomized both on earth and in microgravity. The investigation will keep constant at least one set of parameters  $r$ ,  $ne$ , and  $p$  to compare ground-based and microgravity runs as well as to quantify the effects of gravity-induced convection thereon. The work will correlate process parameters to terminal velocities  $v$  and subsequently to surface properties such as slip and molecular reflection.

### **(2) Falling sphere behavior in a binary gas which maintains a**

### **concentration gradient**

Samples of a given material, either liquid or solid, will be atomized both on earth and in microgravity. The test cell will hold a simple binary gas. Prepared using inert gases of differing molecular weights, this mixture both will diffuse during the course of an experimental run and also will serve as the viscous medium resisting a sphere's free fall. For the first time using the accuracy of microgravity, this setup will demonstrate and assess directly the role of binary gas diffusion on surface interactions. Ground research and theory indicates that the falling sphere will migrate in the direction taken by the diffusion flow of the heavier component, thus quantifying the so-called diffusive slip.

### **(3) Falling sphere behavior in viscous air which maintains a temperature gradient**

Samples of a given material, either liquid or solid, will be atomized both on earth and in microgravity. The test cell will keep a thermal gradient directed transverse to the electric plates. This gradient will serve as the background viscous medium resisting a sphere's free fall. For the first time using the accuracy of microgravity, this set up will demonstrate and assess directly the role of thermal diffusion on surface interactions. This proves particularly well-suited to microgravity work, since the absence of gravity eliminates the effects of convection owing to density gradients. Ground research and theory predict that the falling sphere will migrate in the direction taken by the thermal flow of the hotter portion of the test cell, thus quantifying thermal slip and the radiometric effect.

### **C) Scientific and Technical Significance**

When measured with the high accuracy expected in microgravity, reliable values found for slip coefficients can be expected to resolve fundamental questions

concerning the reflection and absorption of gas molecules. In particular, outstanding questions suggest that the commonly assumed version of the no-slip boundary conditions may require amendment. Further, by finding the coefficient of diffusive reflection, gas dynamics can be understood on a molecular scale. One of two mechanisms, either specular or diffusive reflection, will dominate the interface. Each type of molecular reflection would show differing transport properties and velocity distributions expected in the transition layer linking the solid or liquid to the bulk gas.

If slipping of any kind occurs between the medium and the drop, then the actual terminal velocity will exceed that expected from the simple Stokes' law. This greater measured velocity makes for a larger measured electrical charge and consequently introduces a correction equal to forty percent of the known fundamental charge. This correction to Stokes' law appears most dramatically as drop radii approach six-tenths of the mean free path length. Typical slip coefficients take on values equal to the mean free path of the gas [1]. Typical values reported for oil drops suspended in different gases predict that eighty to ninety percent of gas molecules obey specular reflection at the surface [22]. This varies with the nature of the reflecting surface, as liquids tend to show more diffusive reflection than solids. If microgravity results confirm these sensitive experimental findings, then kinetic theory can rely on several firm assumptions: 1) a fraction  $f$  of the total gas molecules are absorbed on a surface and subsequently reemitted as a Maxwell-Boltzmann gas equilibrated to surface temperature  $T_0$ ; 2) since the gas mean free path varies inversely to pressure, finite slip velocities will dominate surface physics at low pressure; 3) as theory moves from Poiseuille flow to molecular flow, it must displace all surface boundaries a distance equal to the slip coefficient and extrapolate parabolic profiles to zero velocity taken at the new imaginary boundary; 4) new, previously unquantified gas currents can arise in the complete absence of convection--currents which originate from the diminished friction and tangential stress expected for slipping interfaces.

Finally terminal velocities for falling spheres would bear directly on estimating

settling rates for cosmic dust and condensed vapors typically found in the upper atmosphere. Microgravity results could simulate the typical convection found for these high-atmospheric particles. Likewise microgravity would provide the first reliable data collected for falling solids, a finding that would hold great relevance to condensed matter astrophysics.

#### **D) Facilities and Equipment**

The apparatus is self-contained. It measures  $7 \times 7 \times 7 \text{ cm}^3$  and should weigh under ten pounds. It stows easily in a small portion of one middeck modular stowage locker. After removing and inspecting the apparatus, one crewmember will slide it into IML-2's BDP unit along with the test cell and cinecamera. The apparatus is built to perform four tasks: 1) to spray small drops, 2) to suspend them electrostatically between charged plates, 3) to adjust or reverse voltages and 4) to ionize chamber air. Figure VIII, Appendix A gives a summary of expected experimental specifications. When inferred from terminal velocities, the result of surface effects will complete data collection. After flight, the investigator will analyze video records and plot drop trajectories. Using these, he will calculate terminal velocities, electrical charges and corrections to Stokes' law.

The experimental set-up must allow a crewmember to adjust power to the electric plates and ionize air in the test cell\*. Each video exposure will show real-time on film. Data reduction relies exclusively on relative times, such that the known film speed (400 frames per second) will give resolution accurate to within  $2.5 \times 10^{-3}$  seconds. The cinecamera must remain rigidly mounted such that the test cell stays in focus throughout the course of time recording. The plate separation should fill the film frame. These experiments will require no telepresence or telerobotics in Spacelab. The test cell should maintain an ambient temperature varying no more than  $+1^\circ \text{ C}$ . All

these specifications meet current BDPU planning or proposed minor modifications. Modifications mentioned in the proposal carry an asterix. Figures IX and X, Appendix A show a schematic of these modifications.

### **E) Orbital Crew and/or Payload Specialist Training Requirement**

Following launch, the experiment will begin. The slide-in unit to the BDPU will be inserted into the shuttle locker. The crewmember will load one of several stowed canister, primed before flight and filled with liquid either for direct atomizing or for atomizing and subsequent solidifying in situ. At ambient temperature, the latter case will solidify to form small spheres. The crewmember will ensure that both the cinecamera and shielded radium source sit properly on the unit and function as prescribed. He/she will note initial BDPU parameters: temperature, pressure, and voltmeter readings. The crewmember will voice-record comments on experimental conditions. Typical conditions worth noting include accelerations or anomalies that might affect drop trajectories. When combined with the investigator's knowledge of chamber effects (such as transients, power surges, etc.), a transcript of the crewmember's comments will aid post-flight review and analysis of results. The BDPU's cinecamera will record the visual appearance of each drop's trajectory and mark experimental times. The BDPU's background illumination will light the test cell. Suspended inside the test cell, each drop will appear as a well-defined star.

A sequence for each experimental run will commence. The crewmember will initiate this sequence manually. He/she will begin by atomizing a drop cloud. He/she will turn on the BDPU's electric field to a constant and prescribed initial value, then adjust the potential difference until a drop is singled out. The crewmember will center this drop in the viewing area. He/she will voice-record this event as a drop capture. For the length of a run, the crewmember will change the drop trajectory manually. The trajectory will evolve according to the following prescription: 1) as the

charged drop approaches one of the electric plates, he/she reverses the field; 2) after each pass, he/she records a voltmeter reading ; 3) after filming several passes through the viewing area, he/she turns on the ionization source and recharges the dry air filling the chamber; 4) to allow the drop to pick up an ion, he/she turns off the electric field for an instant; 5) beginning again at step one, he/she repeats the sequence using the newly recharged drop. Responding to either a drop leaving the viewing area or to a planned and prescribed change in drop radius, the crewmember will atomize another cloud and select a new drop. Again, he/she signals this change by voice-recording a drop capture.

For planning tests to be conducted in flight, current estimates provide fifteen minutes set-up time, a half-hour run, and fifteen minutes break-down time. Two runs would prove optimal. This will require no special shuttle pointing or positioning and no unusual communication, tracking or orbital needs. Other than the BDPU's existing capability to light the test cell, no special shuttle lighting will prove necessary. While the cinecamera is filming a drop trajectory, accelerations arising from control thrusters will render that particular pass of no significance. However, such an impulse would not present any catastrophic problems: a new drop can always be atomized. Since each pass runs for a short time and the cinecamera records displacements instantaneously, the two-hour period requiring low gravity ( $10^{-6}$  g or better) need not be continuous. (Instantaneous here means that when recorded during a single pass, each drop follows a trajectory taken independently of its previous passes or acceleration history). Therefore as long as each individual pass takes place during low-gravity, a longer sequence including several experimental runs can be scheduled during shuttle maneuvers. Once the crewmember installs the slide-in unit, air stagnancy should follow shortly thereafter. It is estimated that several minutes without residual acceleration should give consistent data. By turning off the electric field and watching for drop drifts, he/she can demonstrate stagnant conditions.

Using the BDPU's cinecamera, the crew will collect all real-time data for the



experiment. It will not need any real-time ground support nor special ground skills or equipment. The investigator should be available to answer questions.

The investigator will analyze this data taken for fixed displacements  $\Delta x$ . He will calculate mean terminal velocities  $v$  from all photographically recorded times  $t$ . Before launch, the investigator will calibrate the cinecamera lens to demark distances and measure radii  $r$ . (N.B. If sufficient time proves available in flight, one additional pass can correlate and calibrate small radii ( $l/r \rightarrow 1$ ) precisely against Brownian motion recorded in the absence of applied electric fields  $E$ .) Chamber parameters such as pressure  $p$  and temperature  $T$  allow the investigator to extract information on mean free paths  $l$  and air viscosity  $\eta$ . These parameters complete the data gathering to find and define surface effects in solids and liquids.

## **IV. Conclusions**

The proposed work calls for experiments addressing both fluid dynamics and transport phenomena, as well as adding to the physics and chemistry of fundamental phenomena. The design will challenge and test critically fundamental assumptions made in surface physics--assumptions concerning molecular reflection, slip conditions, and particle settling rates. The expected results found in low gravity will enhance understanding and confidence with which these assumptions apply. For the first time, the IML-2 flight opportunity will allow a measurement found independently from buoyancy-driven convection and density uncertainties. In short, it will help quantify the necessary physics required to impose conditions on surface boundaries

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Table 1. Typical Data Collected for a Single Drop

*Negative Drop*

*Distance between inner-plates* = 1.010 cm  
*Distance between plates* = 1.600 cm  
*Temperature* = 24.6 °C  
*Density of oil at 25° C* = .8960  
*Viscosity of air at 25.2° C* = .0001836  
*Drop radius* = .000513 cm.  
*Pressure* = 19.01 cm

| Volts             | Fall sec. | Rise sec. | n | $e_2 \times 10^{10}$ | $e_1 \times 10^{10}$ | No. Obs. |
|-------------------|-----------|-----------|---|----------------------|----------------------|----------|
| ( 25.35<br>-25.48 | 22.8      | 24.34     | 7 | 34.47                | 4.923                | 617      |
| ( 25.35<br>-25.48 | 17.2      | 17.8      | 8 | 39.45                | 4.931                | 1,713    |
| ( 13.10<br>-12.29 | 44.9      | 46.9      | 7 | 34.56                | 4.937                | 5,950    |

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Table 2. Correction of Stokes' Law for Small Radii/ Large Free Path

Temperature = 24.6° C  
 Density of oil at 25° C = .8960  
 Viscosity of air at 25° C = .0001836  
 Plate Factor = 25.35 F  
 Pressure = 19.01 cm

| No. | Velocity cm/ sec. | Radius cm. | $e_1 \times 10^{10}$ | Percent Prob Error |
|-----|-------------------|------------|----------------------|--------------------|
| 1   | .001315           | .0000313   | 7.384                | 6.                 |
| 2   | .001673           | 358        | 6.864                | 4.                 |
| 3   | .02613            | 1523       | 5.293                | .5                 |
| 4   | .06800            | 2421       | 5.143                | .7                 |
| 5   | .08843            | 2815       | 5.103                | .3                 |
| 6   | .1644             | 3876       | 5.050                | .3                 |
| 7   | .2175             | 4447       | 5.046                | .4                 |
| 8   | .4074             | 6104       | 5.033                | 1.                 |

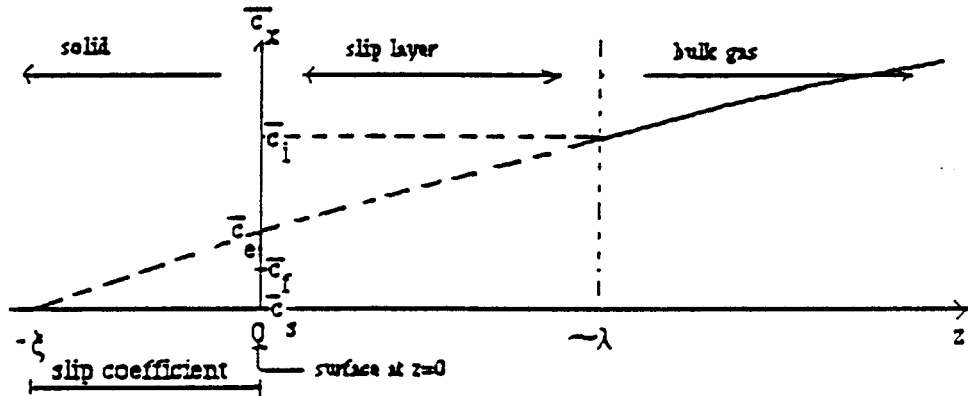
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Table 3.

**Microgravity Advantages to Study Surface Effects**

1. Eliminate buoyancy-driven convection currents
  - A. Particle drifts give 10% error for each .2 mm. of convective length scale
  - B. Gas stagnancy allows the use of smaller spheres, thus:
    1. increasing the accuracy of time measurements
    2. reducing internal convection
    3. allowing one to vary radii instead of mean free path (pressure) and extrapolate Stokes' correction to small slip and infinite pressures
2. Eliminate deviations due to misalignment between gravity and electric field vectors (5 mm. particle drift introduces 1% error).
3. First density-free calculation, which proves important for:
  - A. Solid spheres
  - B. Liquids of uncertain density or composition
  - C. Solids or liquids of uncertain sphericity
4. Allows use of any particle speed (not superimposed gravity acceleration) and thus greatly enhances precision and experimental flexibility

Figure 1. Velocity Profile for Gas Moving Past a Solid Surface



$\bar{c}_e$  = velocity for  $\bar{c}_x$  at the surface by assuming that the functional dependence of  $\bar{c}_x$  on  $z$  in the gas bulk extends all the way to the surface.

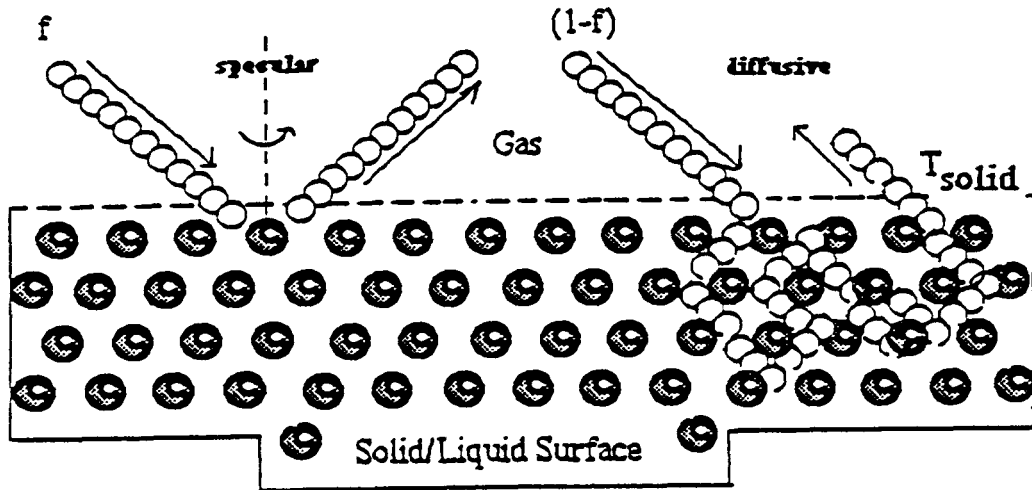
$\bar{c}_i$  = actual velocity for  $\bar{c}_x$  with which the gas atoms strike the surface.

$\bar{c}_f$  = actual velocity for  $\bar{c}_x$  with which the gas atoms scatter from the surface.

$\bar{c}_s$  = velocity of the surface is zero, as would be  $\bar{c}$  if there were no slip; that is, if the flow were continuum.

$\zeta$  = slip coefficient, or jump distance =  $(\bar{c}_e - \bar{c}_s) / (d\bar{c}_x/dz)|_{z=0}$

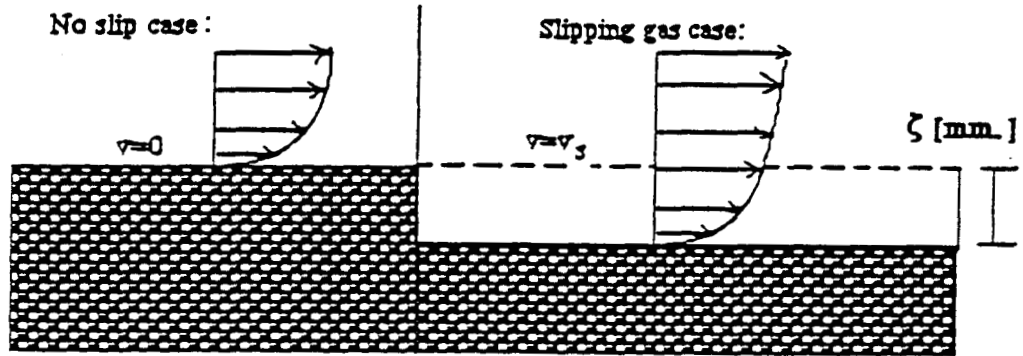
Figure 2. Diffusive vs. Specular Reflection



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Figure 3. Slip Coefficient



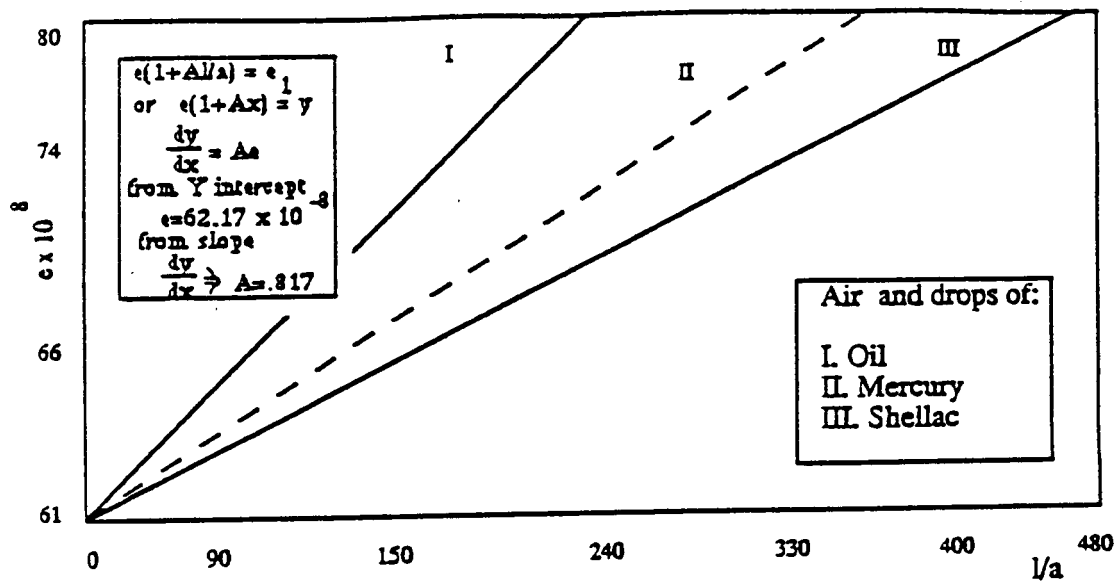
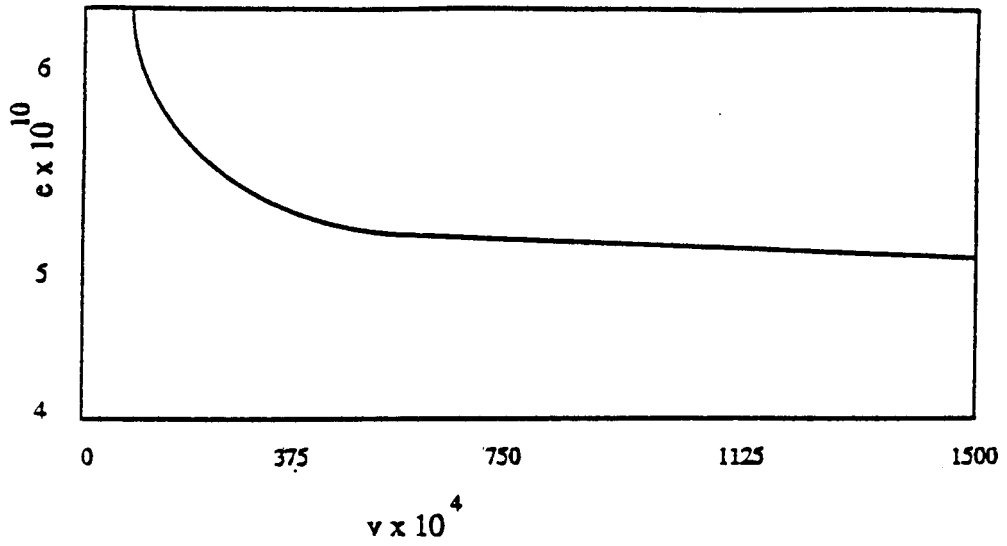


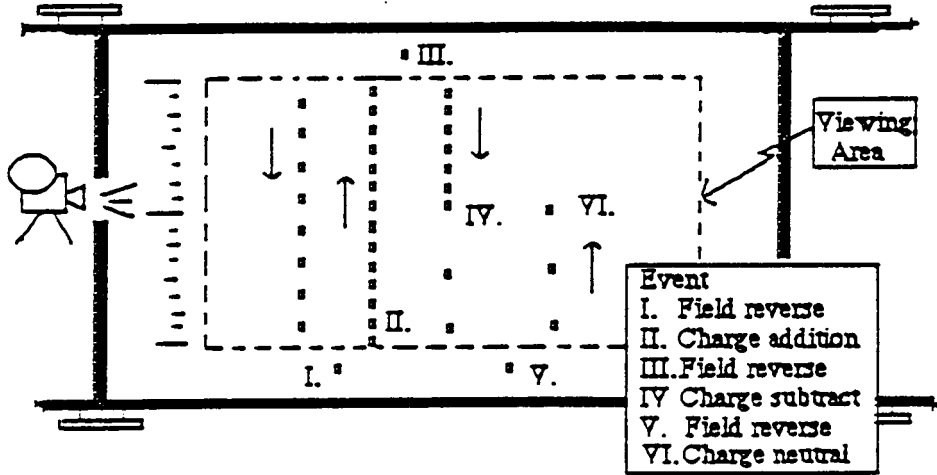
Figure 4. Measured charge as a function of  $l/a$

Figure 5. Low velocity deviation in gravity experiments



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Figure 6. Typical drop trajectories—photographic time measurement



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Figure 7. Circular Plate Schematic and Potential Field

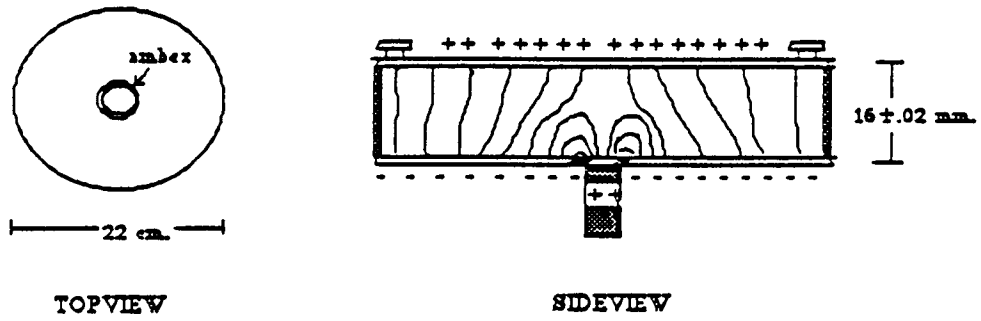
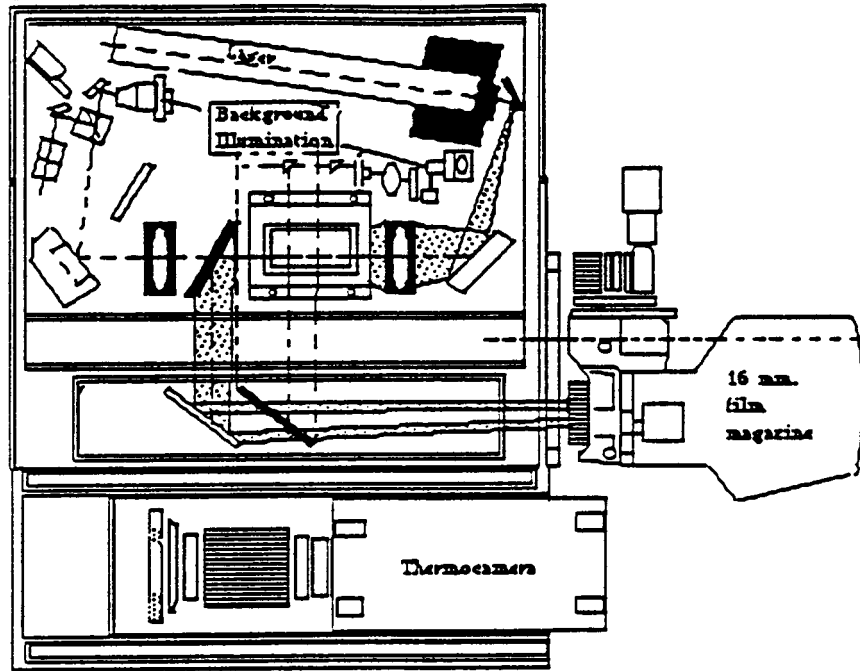


Figure 8. Bubble, Drop and Particle Unit (BDPU)



Technical and Operational Specifications of BDPU

| Parameter                       | ESA Specifications  | Experimental Requirements             |
|---------------------------------|---|---------------------------------------|
| Max. volume                     | 7x7x7 cm. <sup>3</sup>  | 7x7x7 cm. <sup>3</sup>                |
| Field of view                   | 6x4 cm. <sup>2</sup>  | 6x.3 cm. <sup>2</sup>                 |
| Observation systems             | - Cine camera<br>- Still camera<br>- Thermocamera<br>- Interferometer | - Cinecamera                          |
| Illumination                    | - Homogeneous background illumination<br>- Sheet illumination         | - Homogeneous background illumination |
| Injection and detachment system | - Syringes with positioning needles                                   | - Syringes with positioning needles   |
| Sensors                         | Pressure and temperature  | Pressure and temperature              |
| Electrical interface            | 0 → 100 V   | 0 → 100 V                             |
| Humidity                        | <100 ppm H <sub>2</sub> O   | <100 ppm H <sub>2</sub> O             |
| Particulate contamination       | > class 1,000 level   | > class 1,000 level                   |
| Residual acceleration           | <10 <sup>-6</sup> G   | <10 <sup>-6</sup> G                   |
| Time resolution                 | 400 fps   | 400 fps                               |

Figure 9. Bubble, Drop and Particle Unit (BDPU)  
Plus Modifications

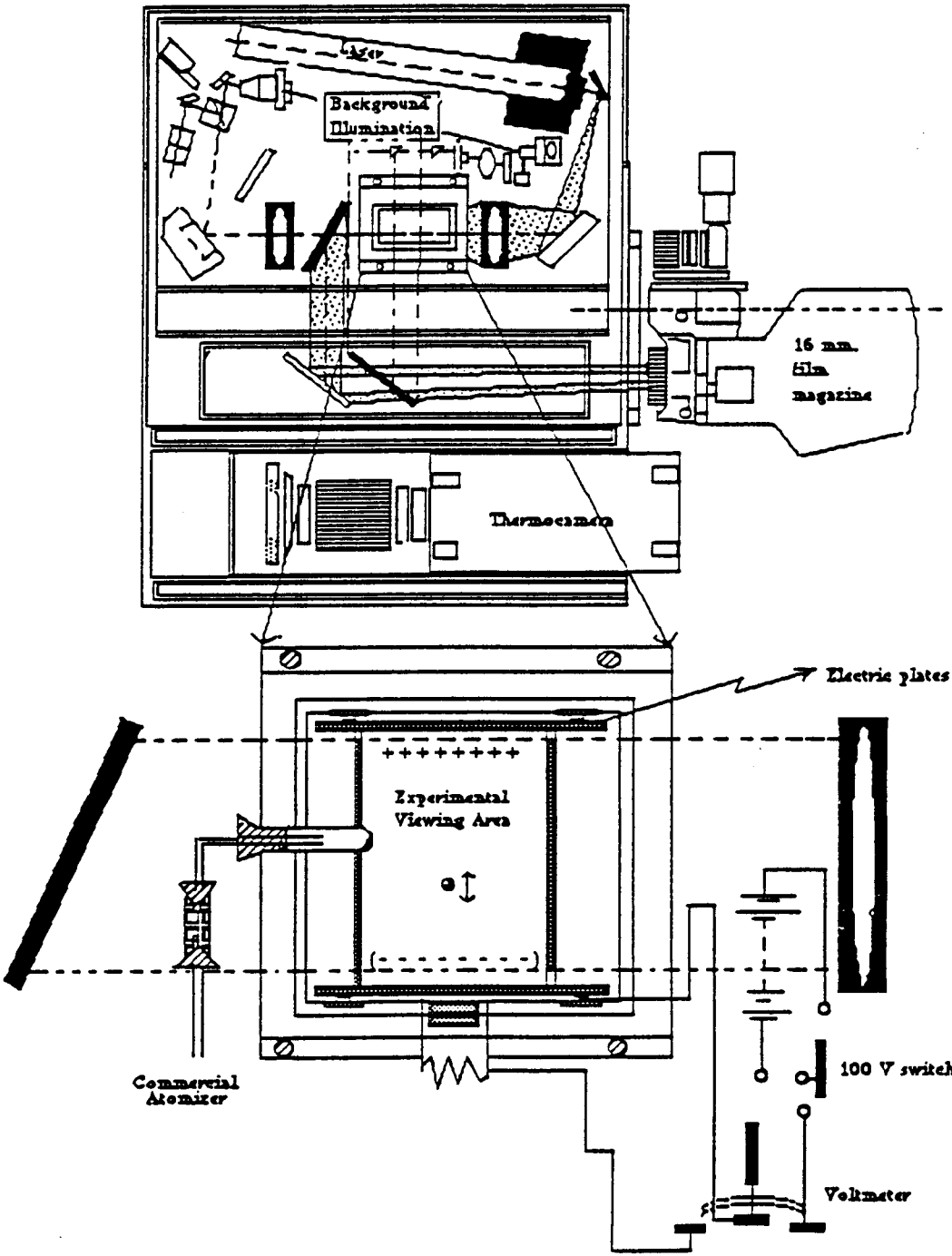
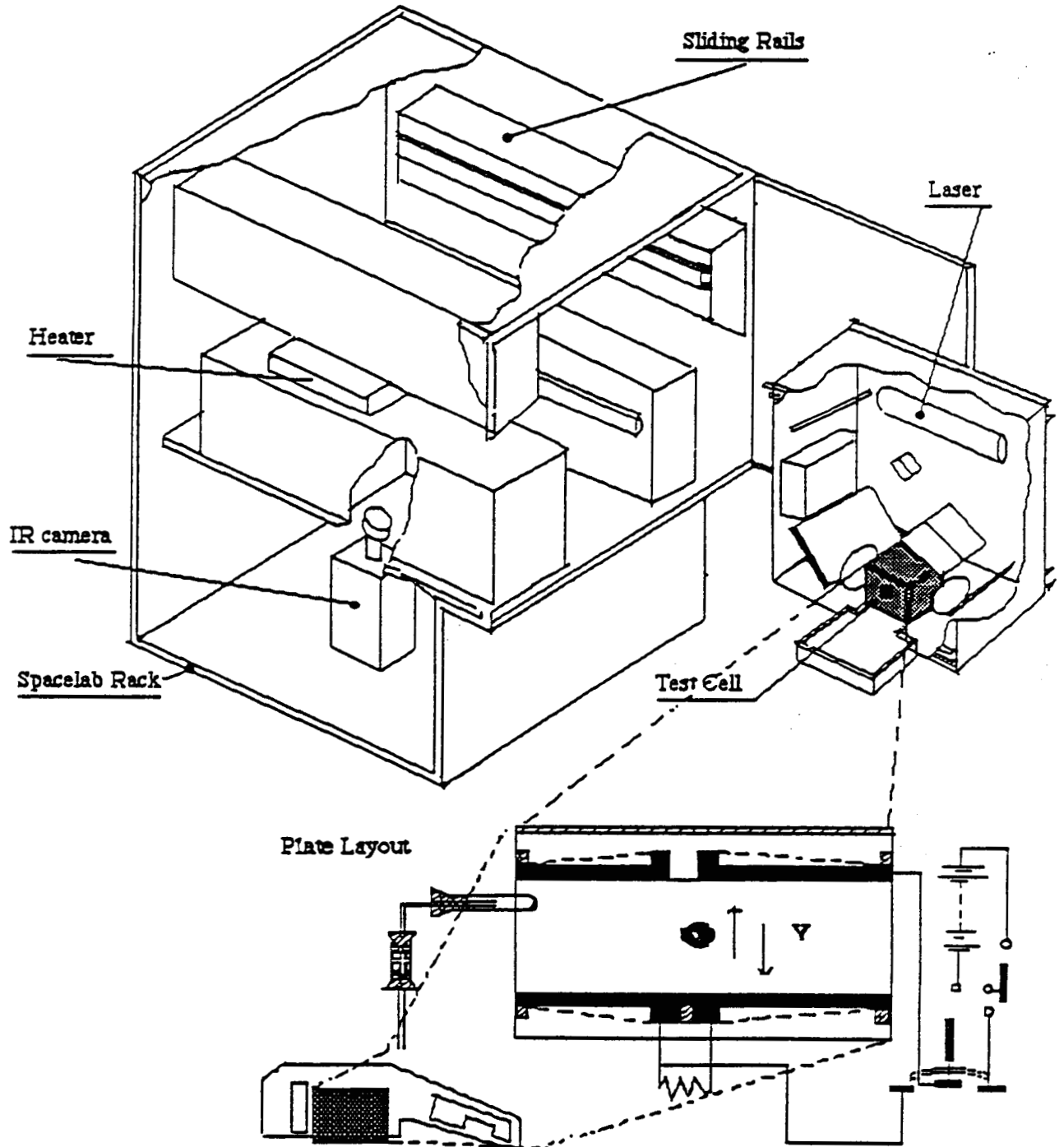


Figure 10. **Modifications To  
Bubble, Drop and Particle Unit / Slide-In Unit**





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| 16. ABSTRACT<br>The investigation describes a proposed experimental program to look at a series of vapor transport properties measured along solid and liquid surfaces. The research objectives proposed are: (1) with accuracy otherwise unobtainable on ground, to determine the coefficient of slip measured between gases and the surface of liquids and solids; (2) for the first time, to classify and tabulate dominant surface effects found for a variety of solids, particularly those crystalized by vapor transport; and (3) to extend understanding of settling rates predicted for cosmic dust and condensed vapor falling through planetary atmospheres.<br>The experimental principle tested is as follows. By electrostatically suspending charged solid and liquid droplets and recording precise times for displacements, surface effects can be understood on a molecular scale. As drop radii decrease, so does the drag force $F$ and the time $t$ required for moving a fixed distance. These changes found in the calculated drag force $F$ give corrections to a Stokes law which otherwise proves valid for large radius spheres. In both its corrected and uncorrected form, it states that the drag force (and in this case, measured electric charge) varies proportionally to the terminal velocity of a falling drop. But unlike the simple Stokes law, the correction is correlated to surface effects occurring on the small drop's surface. These effects are otherwise unmeasurable and include properties such as slip distance and dominant type of molecular reflection. Thus, the micro properties controlling surface physics follow from macro properties such as times and displacements, both found non-intrusively and with great accuracy in microgravity.<br>To an order of magnitude, this method has aided understanding of various liquid-gas interfaces such as oil and water. But to date no similar characterization has proved successful for solids or liquids of uncertain densities. Likewise, no data exist in either ground-based research or as part of a microgravity program that, when collected with the high accuracy expected in low gravity, could definitely settle outstanding questions in kinetic theory, molecular dynamics, and cosmic physics. |  |  |  |  |                       |
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