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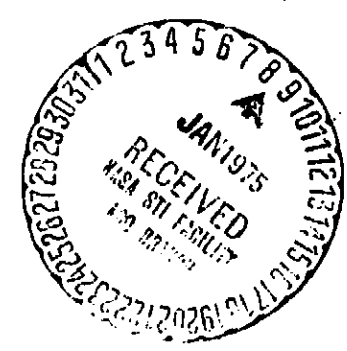
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A THEORETICAL STUDY OF OPTICAL CONTACT OF VITREOUS SILICA

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16. ABSTRACT Optical contact has been proposed as a method of bonding quartz parts of the Stanford relativity Satellite. In this paper the theory of the van der Waals force is outlined and applied to the problem of optical contact. The effect of various contaminations is discussed and a program of experimentation to further study of the problem is presented. REPRODUCIBILITY OF ORIGINAL PAGE IS POOR.			
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1. INTRODUCTION

Optical contact has been proposed as a method of joining fused silica parts in the Stanford relativity satellite. Since little is known about optical contact, we shall present a theory of optical contact and a program of research to further determine the properties of the bond.

When two flat polished glass surfaces are pressed together, an adhesion takes place in the area around the point where the force is applied. The adhered area shows up as a "dark" spot between the glass plates. If the pressure is applied long enough, the spot begins to grow, even when the pressure is removed, until the entire area appears uniformly dark. The glass pieces cannot be separated by force without breaking, indicating that there is considerable adhesion between the surfaces. This phenomenon is called optical contact.

Optical contact has been an obscure and little understood laboratory curiosity for many years, and has been described by many observers, including Lord Rayleigh. Only a complete development of the quantum nature of the forces involved has given us an explanation of this phenomenon. Indeed, any application of optical contact for practical adhesion of glass to glass depends on an understanding of the forces involved.

Optically bonded pieces of quartz can be separated by immersion in a suitable penetrating oil. This is a very useful property allowing parts to be demounted after assembly. In addition to this is the property of distributing strains over the entire surface.

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With mechanical fasteners, the strains are concentrated at the point of fastening. Since the relativity satellite will be cooled to four Kelvins, the spreading of thermally induced strain is an important property.

In this paper we shall present a plausible theory of the nature of optical contact, explain the separation by immersion in liquid, and present a program of experimentation to investigate various parameters that are of technological interest.

11. INTERNAL AND SURFACE STRUCTURE OF FUSED SILICA

It is commonly agreed that all silicates and silicate glasses are made up of silicon oxide tetrahedra which form the basic units of the vitreous skeleton. Structures of this kind follow Pauling's rule, which states that adjacent tetrahedra can combine only at the corners that is, through the common oxygen atoms, but not at the edges or faces. Figure 1 shows a representation of the SiO_4 tetrahedron and the distances involved (1).

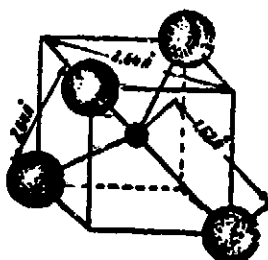


Figure 1. Silicon Oxide Tetrahedron

The internal structure of vitreous silica consists of amorphous chains of the basic tetrahedra (Figure 2). This concept is supported by theoretical work by Zachariasen (2) and x-ray diffraction work by Warren (3).

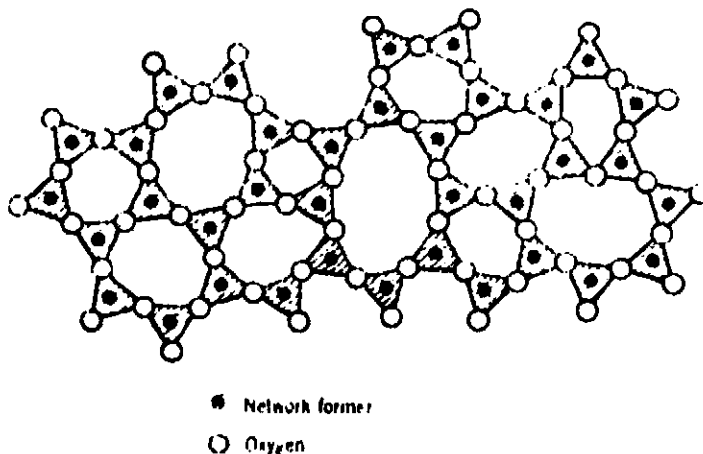


Figure 2. Irregular structure of Vitreous Silica

Each silicon ion is effectively "screened" by the oxygen ions in the arrangement of Figure 2, and the bond between the silicon and oxygen is basically ionic. The structure is not perfectly rigid, since the chains of tetrahedra can flex somewhat with applied force. This means that under sufficient pressure, glass can flow slightly.

In the interior of a piece of silica each silicon ion is surrounded by oxygen ions, thus producing a minimum energy configuration with respect to ionic and overlap bonding. On the surface, however, the silicon ions can possess extra energy since they cannot be surrounded by oxygen ions. Therefore the surface of a piece of silica has excess energy compared to the interior of the same piece, a concept confirmed by measuring the energy necessary to create a new surface by shattering. Since the surface is not in a minimum energy configuration, it follows that a silica surface will be chemically reactive with surrounding matter. For example, a surface silicon ion will possess less energy when bonded to a water molecule than when unbonded, allowing the hydrogen ions to surround each silicon ion in the surface and producing a minimum energy condition similar to the interior.

A silica surface is particularly reactive with another silica surface. When glass bottles come out of the mold, they must be sprayed with a coating or they will seize on contact. The adhesion of silica to silica lowers the surface energy to that of the interior, making a strong bond.

If the silica surface is exposed to water, the hydrogen ions in water serve to screen the silicon ions, forming a chemical bond between the silica and a thin layer of water on the surface.

This layer is at most a few molecules thick; for once silicon ions are screened, the chemical attraction for more water molecules is lessened.

Finally, the surface of a piece of silica exposed to the air for _____ any length of time is covered with a chemically bonded layer of water. Since the water layer forms a barrier to a silica-silica bond, what force is responsible for a optical contact?

III. THE NATURE OF THE BOND

Van der Waals Forces

It is known that there are attractive forces acting between neutral atoms or molecules which are separated by distances that are large compared with their own dimensions. These are known as van der Waals forces and are of a long-range nature, for they decrease according to a power law. It is these van der Waals forces that are responsible for optical contact. We shall give a brief outline of the theory of these forces and present an equation for the binding force between flat plates which takes into account the material of the plates, the temperature, and other parameters. A rigorous derivation of the force equation is given in the appendix.

The Lifshitz theory of the van der Waals forces (5) is based on the idea that the interaction takes place through a fluctuating electromagnetic field. Because of thermodynamic fluctuations, this field is always present in the interior of a material medium and arises from a statistical displacement of the atoms from their equilibrium positions.

The van der Waals forces arise from electromagnetic interactions. As was shown by London in 1930, they arise from second-order perturbation theory applied to the electrostatic interaction between two dipoles; the energy of the interaction is proportional to the inverse sixth power of the distance between them. The existence of attractive forces between neutral atoms suggests analogous forces between any two macroscopic bodies whose surfaces are separated by very small distances.

We can roughly visualize these forces in the following way. The average distribution of electronic charge about a molecule is symmetrical, for this is the minimum energy configuration. However, the electrons move about, so instantaneously the configuration departs from symmetry, and a small dipole exists. This momentary dipole will effect the electronic distribution of a nearby molecule, inducing an oppositely oriented dipole moment onto the second molecule. Although the momentary dipoles and induced dipoles are constantly changing, the net result is attraction between the molecules.

The calculation of these forces on the basis of interaction between individual atoms is impossible, however. In material bodies the close packing of the atoms materially changes the properties of their electronic envelopes, and the presence of matter between the interacting bodies alters the electromagnetic field through which the interaction is effected.

The interaction between atoms in a solid may be treated theoretically by replacing the lattice surrounding the atom in question by a hypothetical effective field, which sums up all the interactions of all the surrounding lattice. Then the interactions between a given atom and the lattice are calculated by considering the interaction between the atom and the effective field, a much simpler calculation. Lifshitz (5) used this approach in his theory of the van der Waals force between macroscopic bodies. He made the following assumptions: 1) the material bodies are continuous media, 2) the interaction is through an effective field. 3) the attractive force is due to the increment to the energy of a material body due to long wavelength fluctuation of this field, and 4) the distance between the bodies must be small compared with the wavelength of the fluctuations of the field.

Using these assumptions Lifshitz arrived at an expression for the force between two plates that varies as the inverse fourth power of the distance. The limitations on the equation are that all characteristic dimensions of the system must be large compared with interatomic spacing.

It should be made clear that the fluctuations in the field manifest themselves externally as quanta of radiation, and wavelengths must be large compared to atomic spacing. All properties of the long-wavelength fluctuations are completely specified by the complex dielectric permeability of the body.

The Force Equation

Dzyaloshinskii, Lifshitz, and Pitaevskii (6) have derived the following equation for the force per unit area F between two solids (See Figure 3):

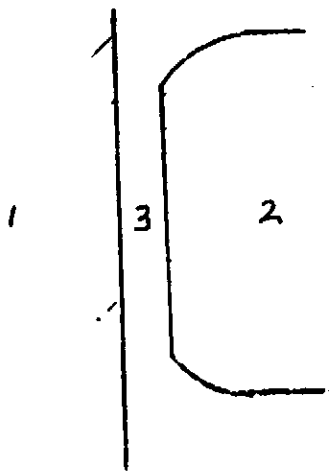


Figure 3. Geometry of Derivation.

$$F = \frac{3\hbar c}{16\pi^2 l^4 (\epsilon_{30})^{\frac{1}{2}}} \int_1^{\infty} \left[\frac{(S_{10} - p)(S_{20} - p)}{(S_{10} + p)(S_{20} + p)} \right.$$

3.1

$$\left. + \frac{(S_{10} - p\epsilon_{10}/\epsilon_{30})(S_{20} - p\epsilon_{20}/\epsilon_{30})}{(S_{10} + p\epsilon_{10}/\epsilon_{30})(S_{20} + p\epsilon_{20}/\epsilon_{30})} \right] \frac{dp}{p^2}$$

where l is the separation between the solids, c is the speed of light in vacuo, \hbar is Planck's constant, ϵ_{n0} the static dielectric constant of the material n ,

$$S_{10} = \left(\frac{\epsilon_{10}}{\epsilon_{30}} - 1 + p^2 \right)^{\frac{1}{2}}$$

3.2

$$S_{20} = \left(\frac{\epsilon_{20}}{\epsilon_{30}} - 1 + p^2 \right)^{\frac{1}{2}},$$

and p is a parameter involving the surface geometry of the integration and is dimensionless.

If we assume that materials 1 and 2 are the same, and material 3 is a vacuum, then $\epsilon_{10} = \epsilon_{20} = \epsilon_0$ and $\epsilon_{30} = 1$.

Equation 3.1 becomes

$$F = \frac{3\hbar c}{16\pi^2 l^4} \int_1^{\infty} \left[\frac{(s-p)^2}{(s+p)^2} + \frac{(s-p\epsilon_0)^2}{(s+p\epsilon_0)^2} \right] \frac{dp}{p^2}$$

3.3

where $S = (\epsilon_0 - 1 + \rho^2)^{\frac{1}{2}}$. This equation is limited by the requirement that $\lambda kT/hc \ll 1$. (For a more complete derivation of this equation, see the appendix).

It is to be noted that equations 3.1 and 3.3 are independent of temperature. This means that for temperatures normally encountered (from roughly four Kelvins to several hundred Kelvins) the temperature variation in the force is very small.

Applications to Quartz

The case of quartz has certain peculiarities because of the specific properties of its absorption spectrum. Quartz absorbs strongly in the ultra-violet (from about 1500 Å) and in the infrared (from several μ) and between these wavelengths is transparent. The contribution of the ultra-violet absorption band to F can be estimated from equation 3.3 by putting ϵ_0 equal to the square of the refraction index in the transparent optical region. The contribution of the infrared is smaller by a factor $h\omega_0/c$ (ω_0 is the infrared absorption frequency); thus we can neglect it and use equation 3.3 to calculate the force.

In Table I we list the characteristic constants for quartz to be put in equation 3.3 for the force calculations.

TABLE I

Values of Physical parameters

ϵ_0	=	3.75	for quartz
ϵ_0	=	80	for water
h	=	1.054×10^{-34}	Joule-Sec.
c	=	3×10^8	m/sec
λ		varies from 10 Å to 10,000 Å	

The force equation (3.3) was then integrated numerically over a range of 10^{-2} to 10^0 . Above 100 the increments to F were converging rapidly to zero ($< 10^{-6}$ parts per integration). The results of this integration and the force per unit area as a function of plate separation are given in the appendix. The values of the force for pure dry quartz range from 2.7×10^{-2} nt/cm² at 100 Å distance to 274 nt/cm² at 10 Å. This force implies considerable adherence.

IV. OTHER ASPECTS OF THE OPTICAL BOND

Effects of Surface Contamination and Surface Roughness on The Bond

Surface contamination and surface roughness can have a large effect on the bonding of two pieces of silica. If particulate contamination is present, then the surfaces are prevented from coming into the intimate contact necessary for optical bonding. Also, if surface irregularities are present, the surfaces will not contact in a large enough area to form a bond.

In an experiment to determine the extent of particulate contamination of "clean" glass, Bastow and Bowden (7) report that when a clean plate, optically polished to within .25 micron of flatness, was lowered onto a similar plate in air, the two surfaces remained about four microns apart and did not come into contact. This was due to the use of ordinary cleaning procedures which left particles about this size on the surfaces. However, when extreme care was taken in cleaning and protecting the surfaces, the distance between the surfaces was about 0.45 microns.

If we assume that two plates are 0.45 micron apart, we can use the theory of the previous section to calculate the van der Waals force. Referring to Table A1 in the Appendix, the value of the force is 6×10^9 nt/cm², a very small force. Therefore, the plates must be pushed together with considerable force in order to bring them close enough together to cause adhesion. This force will increase as the amount of dust contamination increases.

An estimate of the force necessary to push the plates together may be made by the following technique. If there are five particles each five microns in diameter per square centimeter on the surface of a piece of quartz, we may use a method developed by Holm (8) to determine that it takes approximately 100 grams of force to push another plate into contact with the first piece of silica and bury the particles into the silica. This is assuming a contact hardness of 70,000 Kg/cm² for silica. (9).

If the surfaces are not perfectly flat, then the contact area will be quite small. The actual contact area can be calculated using another method of Holm. The load P and the hardness H are related to the contact area A by the formula

$$P = KIA \qquad 4.1$$

where K is 0.02 for a polished surface.

The factor K varies with the degree of flatness of a surface; for a one quarter wave flat the factor is less than .001. If we assume a value of 0.02 then we may calculate the contact area as a function of load.

Table 11 shows the results of this calculation of contact area as a function of varying pressure, neglecting the van der Waals forces.

TABLE II.

Load, K g	Contact Area, cm ²
1	7×10^{-4}
2	1.4×10^{-3}
10	7×10^{-3}
50-	3.5×10^{-2}
100	7×10^{-2}

If we assume that the areas in contact are closer than twenty Angstroms to each other, then from Table II we obtain the figure of 1.4×10^{-3} cm² for a load of two kilograms. This will produce a binding force (See Table AI in the appendix) of over a hundred grams, enough to cause the pieces in contact to seize. This is in agreement with Bastow and Bowden, who reported, "A pressure of several pounds was applied to the top plate The plates now adhered to one another and it required some force to separate them."

We may conclude that the formation of an optical contact bond is a function of the amount of particulate contamination and the surface roughness. If the surfaces are extremely flat, to a quarter or tenth wave, then the factor in Holm's equation is reduced further and bonding will occur at a pressure of a kilogram or less.

The particulate contamination is also an important factor in the formation of the bond, since a large number of particles can prevent the surfaces from coming into intimate contact.

The particulate contamination should be kept as low as possible in order to 1) reduce the force necessary to form the bond and 2) to form a stronger bond. A class 10,000 clean room is recommended as a minimum requirement for assembly environment.

Effects of Surface Water Films

The previous discussion has been based on a clean, bare silica surface. Since a fresh silica surface is extremely reactive, we must assume that any piece of silica which has been ground and polished will have a thin film of water chemically absorbed onto the surface. The presence of water on the surface forces us to change our calculations on the force needed to produce bonding, for McFarlane and Tabor (10) report that the adhesion of a glass bead to a flat plate was dependent on the humidity of the environment. The experiment was repeated with glass plates in a humidity controlled environment and the results are shown in Figure 4.

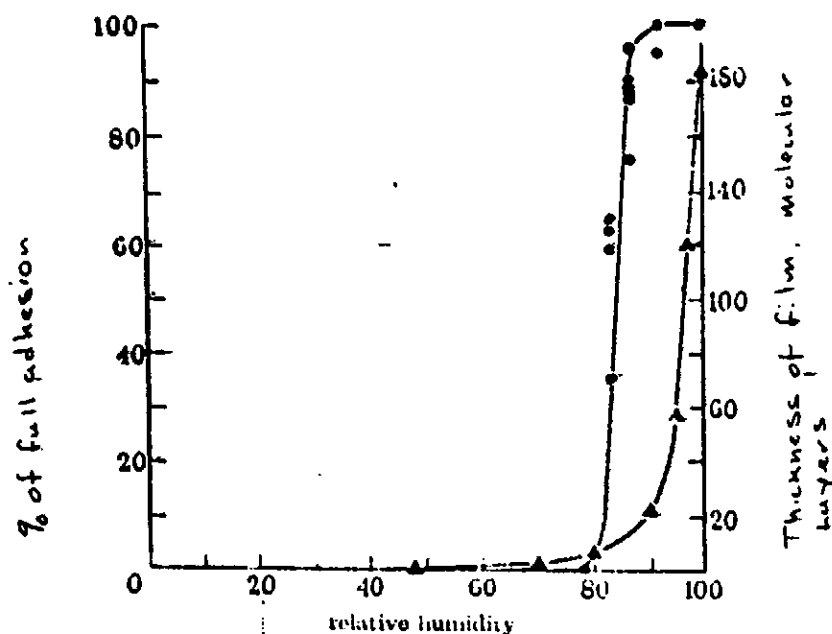


Figure 4. Adhesion of glass surfaces as a function of the humidity of the atmosphere. Curve ● shows adhesion results, Curve ▲ shows the thickness of adsorbed water film as a function of humidity.

It is apparent that the strength of the adhesion depends on the thickness of the water layer on the surface. Let us use the theory of Section III to calculate the force between quartz plates with a thin layer of water adsorbed onto the surface. Using 80 as the value of the dielectric constant, equation 3.3 was evaluated numerically as a function of separation; the results are shown in Table A II in the appendix. For all distances the force is almost five times stronger with a water film than without. These numbers are rough, for there are many unknown parameters. However, this does confirm that the film of water makes a stronger bond than bare silica. More will be said about this in the conclusion section.

The Theory Applied to Other Materials

It is of some interest to determine the van der Waals force between materials other than quartz, and for metals as well. The only material property that enters into the derived force equation (3.1) is the dielectric constant ϵ . For other solid insulators, the force will not vary a great deal from that of quartz, since the range of dielectric constants of inorganic nonmetallic solids is not large.

The basic theory can be applied to metals as well. Assuming that two perfect mirror plates are in vacuum and the dielectric constant approaches infinity, the force equation becomes

$$F = \frac{\pi^2}{240} \frac{hc}{14} \quad 4.2$$

which, for a distance of 10 Angstroms, gives 2×10^4 newton per square centimeter.

The result is much larger than for quartz, giving a strong cold weld to metal.

Methods of Breaking The Bond

It has been determined that the low viscosity oil used to separate gauge blocks can similarly separate optically bonded pieces of quartz. The mechanism of the breaking of the bond was studied by McFarlane and Tabor, (10) who suggest that if materials are bonded through a liquid film, the strength of the bond lies in the amount of energy required to create two fresh surfaces when the bond is broken and the film broken:

That is, energy must be expended to create the new surface. If the pieces bonded together are immersed in a liquid which wets the material, there is no new surface created and the bond strength is lowered to that produced by the viscosity of the liquid. The pieces can be separated slowly, creating no new liquid surface.

Since optically bonded quartz can be separated by the oil, it seems that the optical contact is indeed produced in part by a thin liquid film on the quartz surface. This implies that liquid immersion of some sort may be the best method of separating the plates. Later we shall outline a research program to find the best method of breaking the optical contact.

V. CONCLUSIONS AND RECOMMENDATIONS

From the above theoretical discussion we may draw the following conclusions:

- 1) Optical contact is a complex phenomenon dependent on the previous history of the surfaces to be bonded.
- 2) The bond is a result of van der Waals forces acting between the quartz bodies, but modified by absorbed layers of water on the surface.
- 3) The bond can be broken by immersing the quartz pieces in a suitable liquid and slowly moving them apart.
- 4) The formation of the bond is strongly dependent on surface flatness. However, by applying sufficient pressure, less flat pieces may be bonded. The objects to be bonded must have surfaces flat to one quarter wave; one tenth wave would be better.
- 5) The bond is dependent on the amount of particulate contamination present.

The contact should be initiated in at least a class 10,000 clean environment.

It may be possible to use optical contact in technology; however, much remains unknown about the practical aspects of the bond. The following program of experimental research is strongly recommended to fill gaps in our knowledge of the phenomenon.

Program of Experimental Research

In order to technologically use optical contact, the following areas need further exploration:

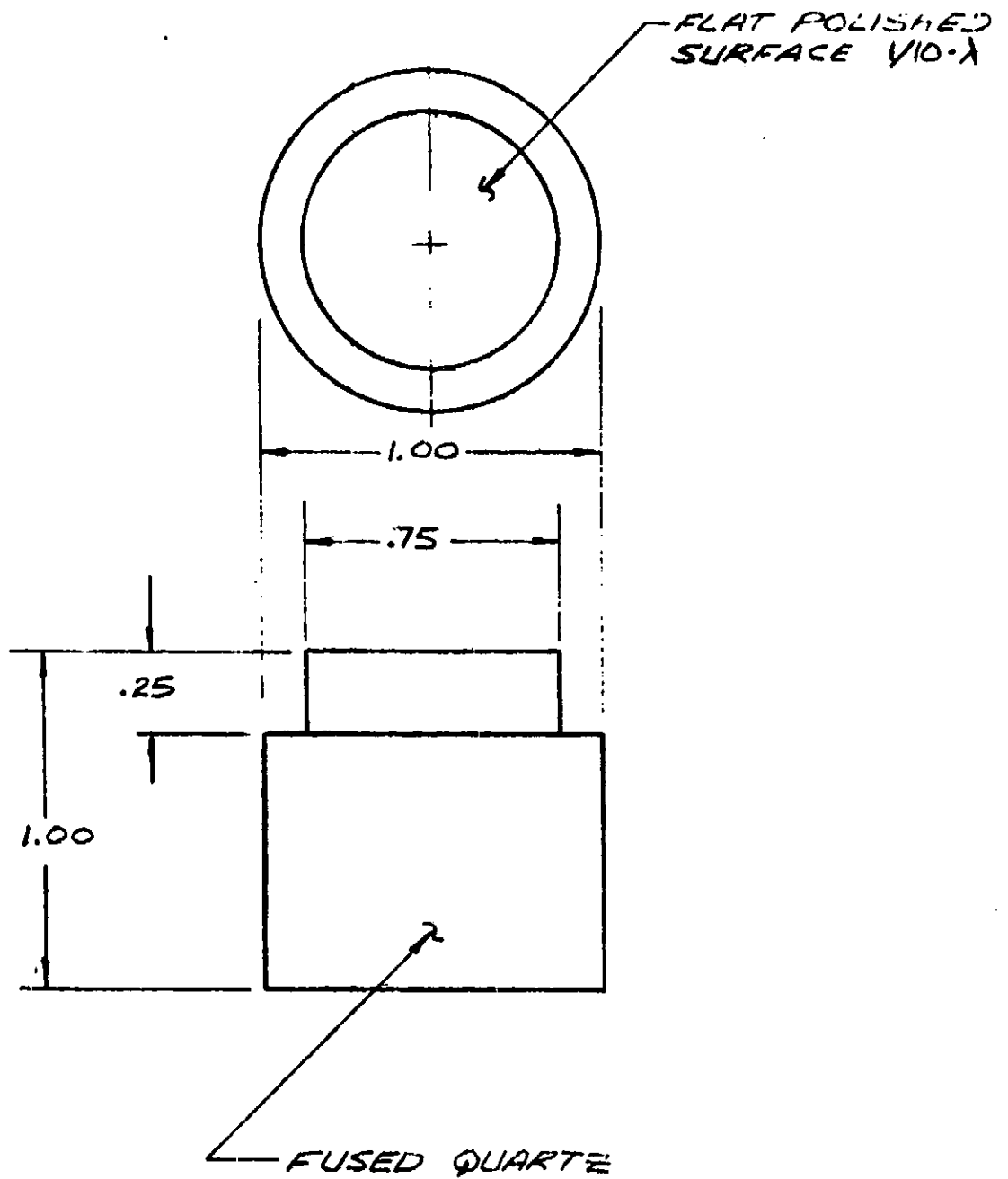
- 1) Pressure necessary to form bond as a function of surface roughness and flatness.
- 2) Effects of surface particulate contamination.
- 3) Effects of humidity on bond initiation.
- 4) Measurement of strength of bond as a function of contamination and amount of water vapor present.
- 5) Strength of bond at cryogenic temperatures, particularly four Kelvins.
- 6) Permanence of bond.

We shall outline experimental methods to implement this exploration.

Material for Experimental Test Pieces

A minimum of five pairs of fused quartz cylinders will be required as test pieces, each with a large height-to-diameter ratio (greater than 1:1), and with mating surfaces polished flat. (Figure 5). These may be obtained by core-drilling sample pieces out of a large flat piece; this will ensure that the test pieces are flat to the edge. The large height-to-diameter ratio will allow the test pieces to be fastened down for strength tests. The pieces should have a neck cut into them to allow attachment to a machine for tensile tests.

Three of the pairs should be finished to flatness of two wavelengths, one half wavelength, and one quarter wavelength, respectively. The other two pairs should be finished as flat as possible, better than one tenth wavelength flat. The latter two will be used for the cryogenic tests.



DIMENSIONS ARE IN INCHES.

Figure 5. Fused quartz test piece for Optical Bonding Experiments

Pressure Necessary to Form Bond

A device to bond the pieces with a known applied pressure is shown in Figure 6. A means of observing the bonding surface while under pressure should be provided.

Humidity Effects

A chamber in which humidity can be easily controlled should be provided for the pressure device. An enclosed area such as the room used for electret research would be an ideal controlled environment, for the humidity can be controlled easily in such a room (Figure 7). The time to form a bond and the bond strength should be measured as a function of humidity under constant load. In addition the load variation at constant humidity should be measured.

Strength Measurements

A tensile stress machine should be provided to pull on the samples and test the bond strength. A commercial machine would serve quite adequately for this purpose. After each bond is formed the force required to break the bond should be measured up to a realistic limit; the force should not be sufficient to break the quartz itself, for the required bond does not have to be that strong.

Basic Lifetime

It has been noted that optically bonded pieces of glass tend to come apart after a certain length of time. Tests should be made to determine

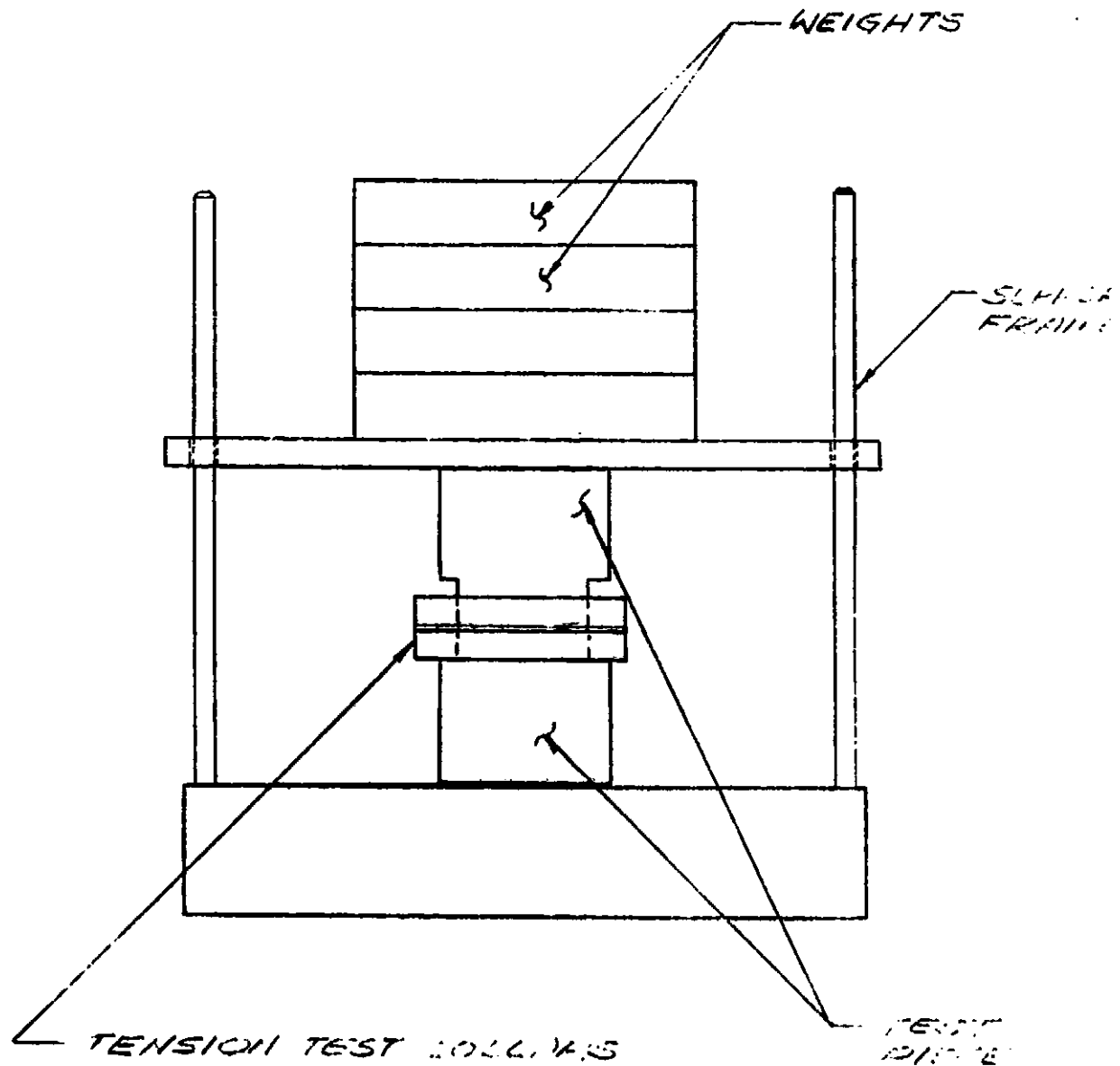


Figure 6. Holding device to apply a known pressure to test pieces to form Optical Bond.

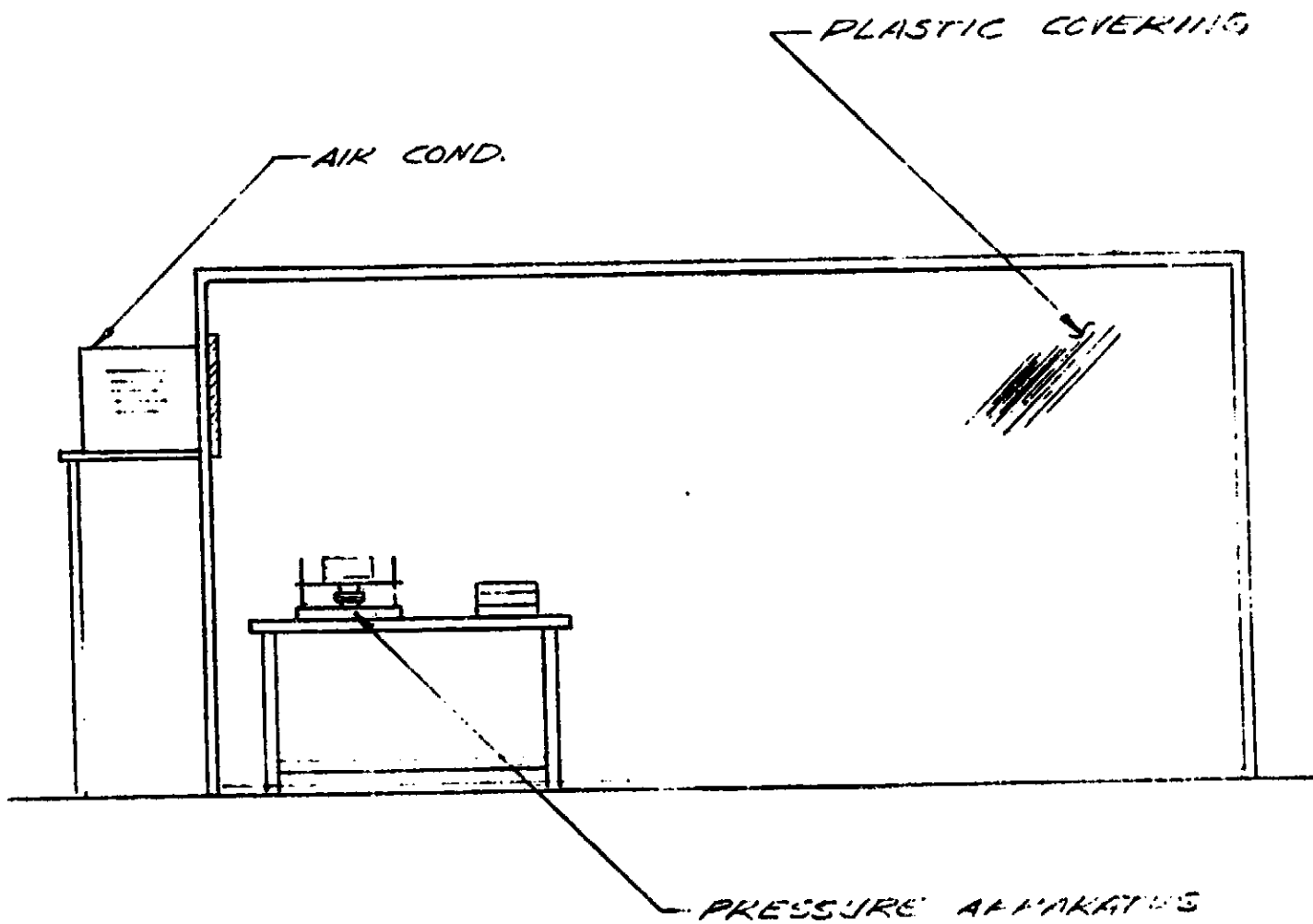


Figure 7. Humidity chamber to test humidity dependence of Optical Bond.

in addition to the tensile strength the decay time of the bond as a function of surface flatness, particulate contamination, and temperature.

Temperature Dependence

A cryostat capable of maintaining the sample at four Kelvins and physically isolating it from the cooling fluid will be required. In addition, the cryostat should allow performance of tensile strength tests while the sample is at cryogenic temperature. Such a cryostat is shown conceptually in Figure 8. _____

Cryostat Description

The quartz test pieces are mounted in a chamber which can be pumped down to at least 10^{-3} torr, and the chamber is sealed with an indium o-ring to exclude the cooling fluid. The test chamber is connected to the top plate which is at room temperature by four stainless steel tubes, each one half inch in diameter by 0.015 inch wall thickness. Another tube will enter the chamber, sealed by bellows, and will exert an upward force on the sample. A needle valve of special design will allow cooling fluid to enter the chamber in a controlled way.

The upper part of the cryostat should consist of a sturdy plate for mounting and a vacuum connection and valve. The tensile stress tube should be attached to a balance beam so weights may be attached to put stress on the sample. (Figure 9).

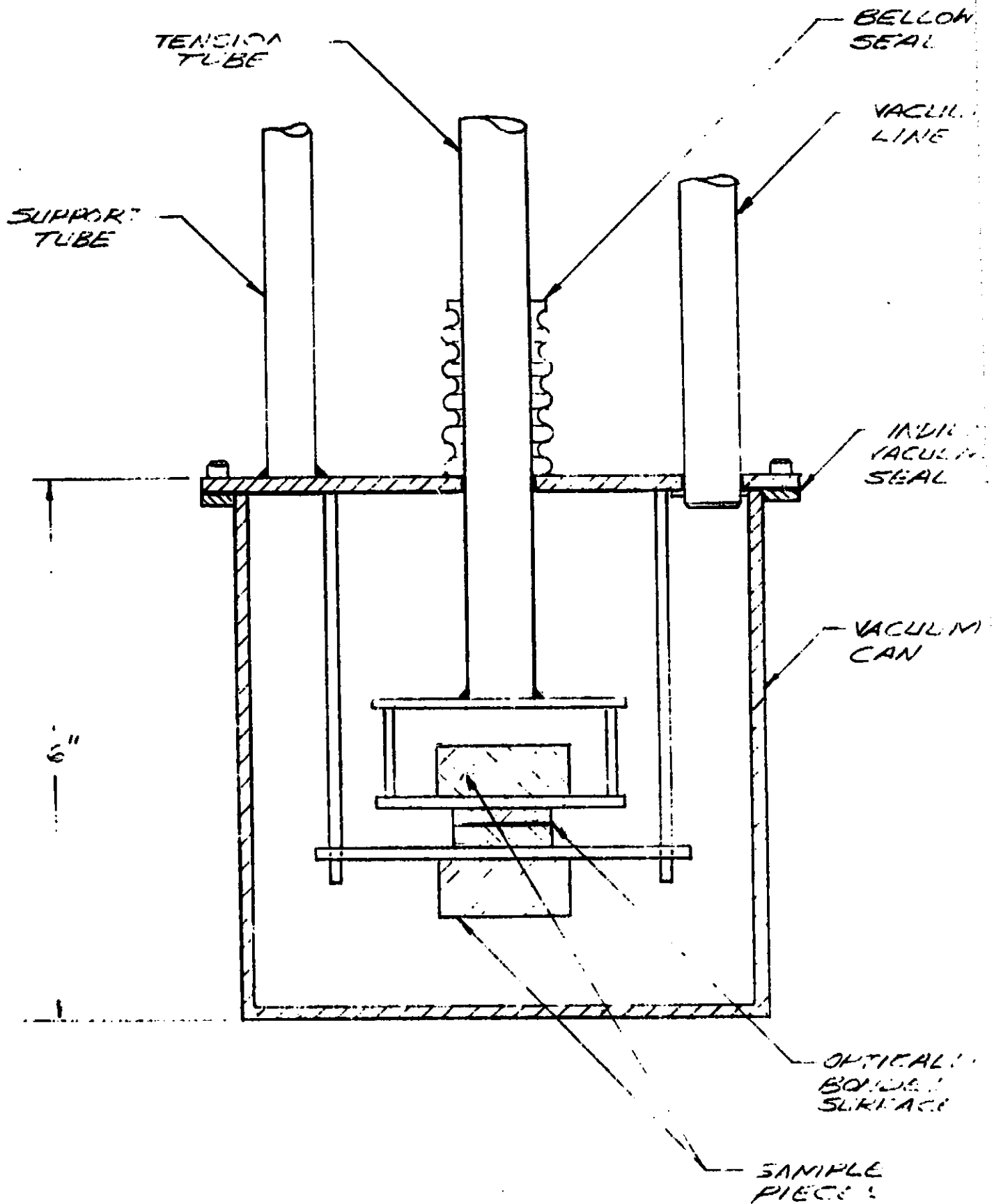


Figure 8. Tensile Test Cryostat Sample Chamber

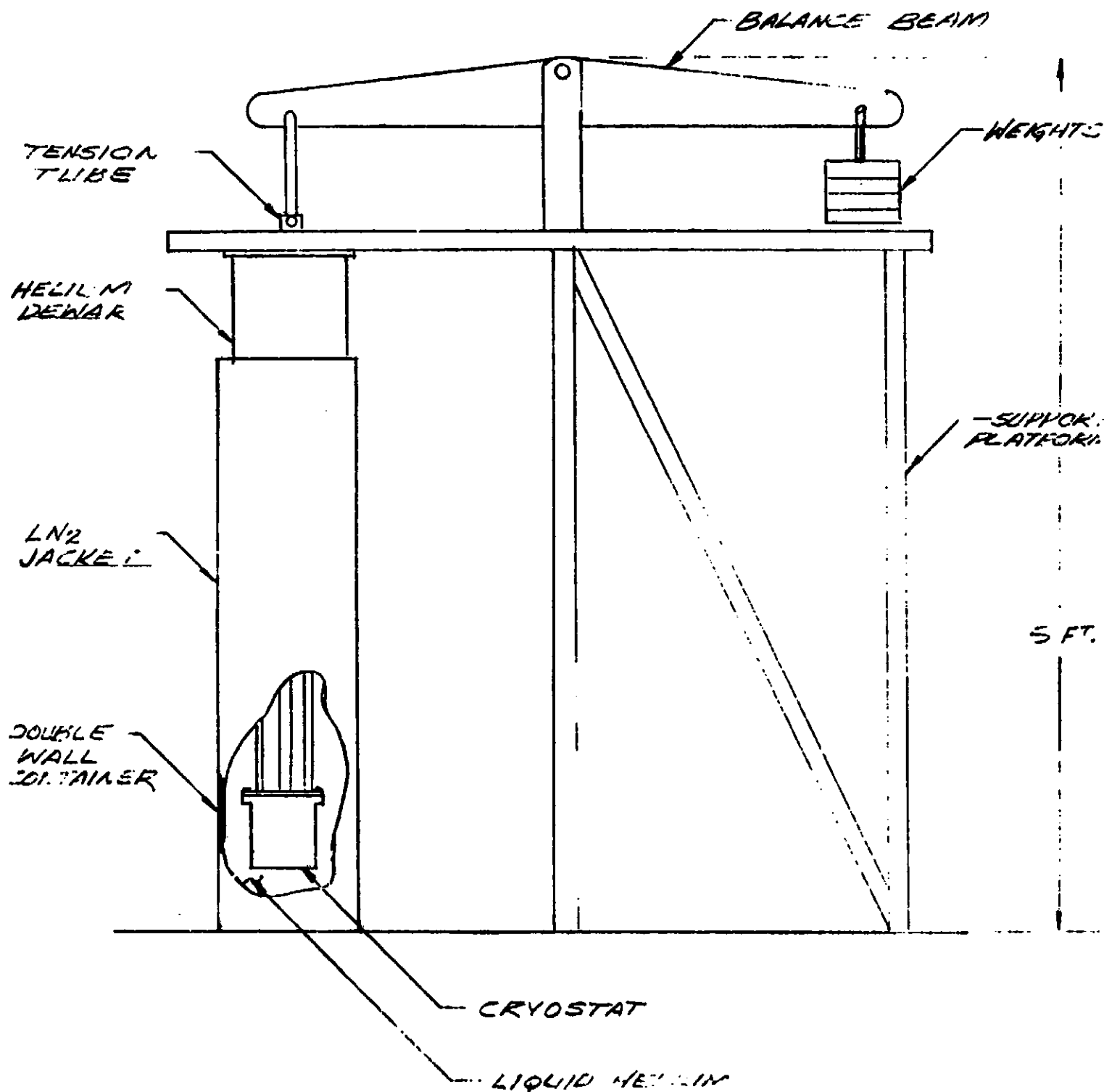


Figure 9. Overall View, Tensile Test Cryostat

The test dewar should be of metal and have two sections, one for a liquid nitrogen jacket surrounding the center where liquid helium may be placed.

The program of experiment should mainly test bond strength at four Kelvins as a function of humidity when the bond is made. Also, the effects of immersion in liquid helium should be tested.

In conclusion this program of experimentation should yield sufficient information to determine the feasibility of using the optical bond on the relativity satellite. Also techniques will be developed to aid in technological use.

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APPENDIX I. DERIVATION OF THE FORCE EQUATION

Appendix 1. Derivation of the Force Equation

Following Dzyaloshinskii, Lifshitz, and Pitaevskii we shall present an outline of the derivation of the force equation 3.1. For more detail the reader is referred to reference 6.

Using the Feynman invariant technique, the above authors solve in principle the calculation of the van der Waals part of the thermodynamic variables of a body in terms of the Green's functions as propagators of the interaction.

The gap separating two bodies may be considered to be filled with any liquid. We shall denote the two solid bodies by the indices 1 and 2, while the medium which fills the gap will be denoted by the index 3. The gap will be represented by two parallel planes.

The total force acting on the body 2 can be calculated as the total momentum flux that enters the body from medium 3, that is, as an integral,

$$\int \sigma_{ik} d f_k$$

over the surface; where $\underline{\sigma}_{ik}$ is the stress tensor in medium 3. From the Feynman techniques the stress tensor can be written in terms of the Green's functions as

$$\sigma_{ik} = \frac{T}{2\pi} \sum \left\{ \epsilon_3 \left[D_{ik}^E(\vec{r}, \vec{r}') - \frac{1}{2} \delta_{ik} D_{ii}^E(\vec{r}, \vec{r}') \right] + D_{ik}^H(\vec{r}, \vec{r}') - \frac{1}{2} \delta_{ik} D_{ii}^H(\vec{r}, \vec{r}') \right\} \quad A1.1$$

where \underline{T} is the temperature, ϵ_3 is the dielectric permeability of medium 3, and the D's are the Green's functions.

We can now write the force per unit area of body 2 as

$$F(l) = \sigma_{yx}'(l) = \frac{I}{4\pi} \sum_{n=0}^{\infty} \left\{ \epsilon_3 [D_{yy}^E(l, l; \xi_n) + D_{zz}^E(l, l; \xi_n) - D_{xx}^E(l, l; \xi_n)] + D_{yy}^H(l, l; \xi_n) + D_{zz}^H(l, l; \xi_n) - D_{xx}^H(l, l; \xi_n) \right\}, \quad A1.2$$

where a positive force corresponds to an attraction and a negative to repulsion.

The problem is reduced to solving the standard equations for the Green's functions. It can be shown that the parts of the Green's functions which depend on the sum $\underline{x} + \underline{x}'$ make no contribution to the expression A1.2 for the force \underline{F} . If we put $\underline{x} = \underline{x}'$ in a solution of the form $f^-(\underline{x} + \underline{x}')$ we would obtain a momentum flux in the gap which varied with the coordinates, and this would contradict the conservation of momentum. Therefore, only those parts of the Green's functions which depend on $\underline{x} - \underline{x}'$ will be taken.

The Green's function solutions are given as follows:

$$D_{zz} = \begin{cases} A \exp(\omega_1 x), & x < 0 \\ B \exp(-\omega_2 x), & x > 0 \end{cases} \quad A1.3$$

$$C_1 \exp(\omega_3 x) + C_2 \exp(-\omega_3 x) = \frac{\lambda_1}{\omega_3} \exp(-\omega_3 |x-x'|),$$

$$\dots x < l$$

Determining constants from boundary conditions we have

$$D_{zz} = \frac{4\pi}{w_3 \Delta} \cosh w_3(x-x'), \quad A1.4$$

where

$$\Delta = 1 - \exp \left[(2w_3 l) \frac{(w_1 + w_3)(w_2 + w_3)}{(w_1 - w_3)(w_2 - w_3)} \right], \quad A1.5$$

where $w = (\epsilon \xi_n^2 + q^2)^{1/2}$

$\xi_n = 2\pi n k T$, and q is a function of the surface.

In like manner we can get for D_{yy}

$$D_{yy} = \frac{4\pi w_3}{\xi_n^2 \epsilon_3 \bar{\Delta}} \cosh w_3(x-x'), \quad A1.6$$

where $\bar{\Delta} = 1 - \exp \left[(2w_3 l) \frac{(\epsilon_1 w_3 + \epsilon_3 w_1)(\epsilon_2 w_3 + \epsilon_3 w_2)}{(\epsilon_1 w_3 - \epsilon_3 w_1)(\epsilon_2 w_3 - \epsilon_3 w_2)} \right]$,

and the other Green's functions:

$$D_{xy} = \frac{4\pi q}{\xi_n^2 \epsilon_3 \bar{\Delta}} \sinh w_3(x-x') \quad A1.7$$

$$D_{xx} = \frac{4\pi q^2}{\xi_n^2 \epsilon_3 w_3 \bar{\Delta}} \cosh w_3(x-x'). \quad A1.8$$

If we now calculate the quantities $D_{ik}^E(x, x'; \bar{q}; \xi_n)$ and $D_{ik}^H(x, x'; \bar{q}; \xi_n)$ and substitute them in eqn. (A1.2), we get:

$$F(l) = -\frac{T}{2\pi} \sum_{n=0}^{\infty} \int_0^{\infty} q dq \cdot \omega_3 \left(\frac{l}{\Delta} + \frac{l}{\Delta} \right), \quad \text{A1.9}$$

Transforming to a new variable of integration ρ , where $q = \xi_n \left[(\epsilon_3(\rho^2 - 1))^{1/2} \right]$ and returning to a normal system of units, we obtain the final expression for the force F acting on unit area of each of the two bodies (media 1 and 2) separated by a gap of width l which is occupied by medium 3

$$F(l) = \frac{kT}{\pi c^3} \sum_{n=0}^{\infty} \epsilon_3^{-3/2} \xi_n^2 \int_0^{\infty} \rho^2 \left\{ \frac{(S_1 + \rho)(S_2 + \rho)}{(S_1 - \rho)(S_2 - \rho)} \exp\left(\frac{\lambda \rho \xi_n}{c} \sqrt{\epsilon_3}\right) - 1 \right\}^{-1} + \left[\frac{(S_1 + \rho \epsilon_1/\epsilon_3)(S_2 + \rho \epsilon_2/\epsilon_3)}{(S_1 - \rho \epsilon_1/\epsilon_3)(S_2 - \rho \epsilon_2/\epsilon_3)} \exp\left(\frac{\lambda \rho \xi_n}{c} \sqrt{\epsilon_3}\right) - 1 \right]^{-1} \right\} d\rho \quad \text{A1.10}$$

where $S_1 = (\epsilon_1/\epsilon_3 - 1 + \rho^2)^{1/2}$; $S_2 = (\epsilon_2/\epsilon_3 - 1 + \rho^2)^{1/2}$; $\xi_n = \sqrt{2\pi n k T / \hbar}$; $\epsilon_1, \epsilon_2, \epsilon_3$

are functions of the imaginary frequency $\omega = \xi_n (\hbar + \epsilon(\lambda \xi))$; and k is Boltzmann's constant. The summation is taken over integral values of n , and the dash attached to the summation sign denotes that the term with $n=0$ is given half weight. Positive values of F correspond to attraction between the bodies and negative values to repulsion.

This formula (1961, p. 4) for bodies separated by an empty gap) was first derived by Lifshitz by another method which did not use the techniques of quantum field theory. These techniques are, however, necessary for the generalization of the result to a gap filled by an arbitrary medium.

The only macroscopic properties of the bodies that determine the strength of the molecular interactions are the imaginary part of their complex dielectric permeabilities. If the bodies are identical ($\epsilon_1 = \epsilon_2$) then the expression under the integral is always positive for all the terms of the sum in (A1.10) and for given values of p and ξ_0 it decreases monotonically as ξ increases. Hence it follows that $F > 0$ and $dF/dl < 0$, i.e. identical bodies attract one another irrespective of the size of the gap between them while the attractive force decreases monotonically as the gap increases.

This statement is also valid for two different media which are separated by an empty gap ($\epsilon_3 = 1$). If the bodies differ and the space between them is filled by some fluid, then the force between them can be either attractive or repulsive (see below).

The general formula (A1.10) is very complicated, but it can be considerably simplified if we use the fact that the influence of temperature on the interaction force usually turns out to be negligible.

Because of the presence of an exponential under the integral in A1.10 the important terms in the sum will be those for which $\xi_n \sim c/l$ or $n \sim ch/kT$. For $l kT/c h \gg 1$, large values of n will thus be important and we can replace the sum by an integration over $dn = (h/2\pi kT) d\xi$. The temperature now drops out of the formula and we are left with the following result:

$$F = \frac{q_1}{2(\sigma_1^2 \epsilon_1^3)} \int_0^\infty \int_0^\infty \rho^2 \xi^3 d\rho d\xi \left\{ \left[\frac{(\epsilon_1 - \rho)(\epsilon_2 - \rho)}{(\epsilon_1 + \rho)(\epsilon_2 + \rho)} \exp\left(\frac{2\rho \xi l}{c}\right) - 1 \right] + \left[\frac{(\epsilon_1 + \rho)(\epsilon_3 - \rho)}{(\epsilon_1 - \rho)(\epsilon_3 + \rho)} \exp\left(\frac{2\rho \xi l}{c}\right) - 1 \right] \right\} d\rho d\xi \quad \text{A1.11}$$

Equation (A1.11) is still complex but it can be simplified further in two important limiting cases.

First we consider the limiting case of small distances, by which we mean distances which are small compared to the wavelengths λ which characterise the absorption spectra of the given bodies. The temperatures which are applicable for condensed bodies are certainly small compared to the $h\nu$ which are important here (for example, in the visible region), and therefore the inequality $kT/h\nu \ll 1$ is known to be satisfied.

Because of the exponential factor $\exp(-2\rho\xi\sqrt{\epsilon_2}/c)$ in the denominators of the expression under the integral, those values of ρ for which $\rho\xi/c \sim 1$ are dominant in the integration with respect to ρ . In this case $\rho \gg 1$ and therefore we can put $s_1 \approx s_2 \approx \rho$ in the main terms. In this approximation the first term in curly brackets in (A1.11) is zero, while the second term gives on the introduction of the variable of integration $x = 2\rho\xi\sqrt{\epsilon_2}/c$

$$F = \frac{h}{16\pi^2 l^3} \int_0^\infty \int_1^\infty x^2 \left[\frac{(\epsilon_1 + \epsilon_3)(\epsilon_3 + \epsilon_2)}{(\epsilon_1 - \epsilon_3)(\epsilon_2 - \epsilon_3)} e^x - 1 \right]^{-1} dx d\xi \quad \text{A1.12}$$

(in this approximation the lower limit of integration with respect to ρ is replaced by zero).

This force is proportional to the inverse cube of the distance, as would be predicted by the usual theory of van der Waals forces between two atoms. The functions $(\epsilon_i - 1)^{-1}$ decrease monotonically as ξ increases and tend to zero. Therefore values of ξ which lie beyond a certain ξ_0 make a negligible contribution to the integral; the condition that l should be small is that $l \ll c/\xi_0$.

To estimate the accuracy of the limiting behavior derived above it is useful to have the next term of the expansion of the function $F(\xi)$. Calculation from the general formula (A1.11) gives (for similar bodies separated by a vacuum, $\epsilon_1 = \epsilon_2 = \epsilon_0$, $\epsilon_3 = 1$) the expression

$$-\frac{\hbar}{8\pi^2 c^2 l} \int_0^\infty \frac{\xi^2 [\epsilon(i\xi) - 1]^2}{\epsilon(i\xi) + 1} d\xi \quad \text{A1.13}$$

which must be added to (A1.12). However, it is not possible to make a realistic estimate of the region of validity of the limiting law without knowing the form of the function $\epsilon(i\xi)$.

To an accuracy which is quite sufficient in practice, eq. (A1.12) can be written in an even simpler form by neglecting unity compared to the term with e^x in the square brackets. (The accuracy of this reduction is connected to the fact that an integral of the form

$$\frac{a}{n!} \int_0^\infty \frac{x^n dx}{ae^x - 1} \quad \text{A1.14}$$

varies very little as a varies from ∞ to 1; from 1 to 1.2 for $n=2$, to 1.08 for $n=3$ to 1.04 for $n=4$, etc.) Then the integration with respect to dx can be carried out in an elementary way, and instead of (A1.12) we get

$$F = \frac{\hbar \omega}{8\pi^2 c^2 l^3} ; \quad \omega = \int_0^\infty \frac{(\epsilon_1 - \epsilon_3)(\epsilon_2 - \epsilon_3)}{(\epsilon_1 + \epsilon_3)(\epsilon_2 - \epsilon_3)} d\xi \quad \text{A1.15}$$

The quantity $|\omega|$ is some characteristic frequency for the absorption spectra of all three media.

We now turn to the opposite limiting case, that of "large" distances; $l \gg \lambda_0$. We shall however suppose that the distances are not so large as to invalidate the inequality $l\kappa T/hc \ll 1$.

We introduce into the general formula (A1.11) a new variable of integration $\chi = 2\rho l \xi / c$, but as the second variable we take not ξ (as above), but ρ :

$$F = \frac{hc}{32\pi^2 l^4} \int_0^\infty \int_1^\infty \frac{\chi^3}{\rho^2} \epsilon_3^{3/2} \left\{ \left[\frac{(S_1 + \rho)(S_2 + \rho)}{(S_1 - \rho)(S_2 - \rho)} \exp(\chi \sqrt{\epsilon_3}) - 1 \right]^{-1} \right. \quad \text{A1.16}$$

$$\left. + \left[\frac{(S_1 + \rho \epsilon_1/\epsilon_3)(S_2 + \rho \epsilon_2/\epsilon_3)}{(S_1 - \rho \epsilon_1/\epsilon_3)(S_2 - \rho \epsilon_2/\epsilon_3)} \exp(\chi \epsilon_3^{1/2}) - 1 \right]^{-1} \right\} d\rho d\chi$$

Because of the presence of the factor $\exp(\chi \epsilon_3^{1/2})$ in the denominators, values of $\chi \sim \sqrt{\epsilon_3} \leq 1$ are the only ones important in the integration over $d\chi$, and since $\rho \geq 1$ the argument of the function ϵ for large l is nearly zero over the whole of the important range of the variables. Because of this we can replace $\epsilon_1, \epsilon_2, \epsilon_3$ by their values for $\xi = 0$ i.e., the electrostatic dielectric constants. If we then replace $\chi(\epsilon_3)^{1/2}$ by χ we finally obtain the following result

$$F = \frac{3hc}{32\pi^2 l^4 (\epsilon_{30})^{1/2}} \int_0^\infty \int_1^\infty \frac{\chi^3}{\rho^2} \left\{ \left[\frac{(S_{10} + \rho)(S_{20} + \rho)}{(S_{10} - \rho)(S_{20} - \rho)} e^\chi - 1 \right]^{-1} \right. \quad \text{A1.17}$$

$$\left. + \left[\frac{(S_{10} + \rho \epsilon_{10}/\epsilon_{30})(S_{20} + \rho \epsilon_{20}/\epsilon_{30})}{(S_{10} - \rho \epsilon_{10}/\epsilon_{30})(S_{20} - \rho \epsilon_{20}/\epsilon_{30})} e^\chi - 1 \right]^{-1} \right\} d\rho d\chi ;$$

$$S_{10} = (\epsilon_{10}/\epsilon_{30} - 1 + \rho^2)^{1/2},$$

$$S_{20} = (\epsilon_{20}/\epsilon_{30} - 1 + \rho^2)^{1/2},$$

where $\epsilon_{10}, \epsilon_{20}, \epsilon_{30}$ are the electrostatic dielectric constants.

In accordance with the above property of integrals of the type of (A1.14), equation (A1.17) can be written with considerable accuracy in the form:

$$F = \frac{3hc}{16\pi^2 l^2 (\epsilon_{30})^2} \int_0^{\infty} \left[\frac{(s_{10} - p)(s_{20} - p)}{(s_{10} + p)(s_{20} + p)} + \frac{(s_{10} - p\epsilon_{10}/\epsilon_{30})(s_{20} - p\epsilon_{20}/\epsilon_{30})}{(s_{10} + p\epsilon_{10}/\epsilon_{20})(s_{20} + p\epsilon_{20}/\epsilon_{30})} \right] \times \frac{dp}{p^2} \quad \text{A1.18}$$

Here only one integration remains and in principle this can be reduced to elementary functions; the result is, however, so unwieldy that in real calculations it is better to use numerical integration.

It was pointed out above that if the two bodies differ and the medium between them is filled with fluid the interaction can be either an attraction or a repulsion. Thus, from (A1.15) it is clear that if $\epsilon_1 - \epsilon_3$ and $\epsilon_2 - \epsilon_3$ have opposite signs in the important frequency range, then $F < 0$, i.e. the bodies will repel each other for "small" separations. At "large" separations the forces are determined by the electrostatic values of the dielectric permeability: if $\epsilon_{10} - \epsilon_{30}$ and $\epsilon_{20} - \epsilon_{30}$ have the same sign $F > 0$, while if their signs differ $F < 0$. Furthermore, since the relative magnitudes of $\epsilon_{10}, \epsilon_{20}, \epsilon_{30}$ are not in general related to the behavior of the functions $\epsilon_1(i\xi), \epsilon_2(i\xi), \epsilon_3(i\xi)$ in the frequency range which is important for these bodies, it is possible in principle to have cases in which F changes sign at some value of l .

The temperature dependence has dropped out in equation A1.18, implying that the force is independent of temperature over the range normally encountered.

This is true as long as $kT \ll \hbar\omega$, for at optical frequencies $T > 10^4$ K. Therefore as long as we are at temperatures less than 1000 Kelvins there is little change of the van der Waals force with temperature.

APPENDIX II. TABLES OF VAN DER WAALS FORCES AS A FUNCTION OF DISTANCE

RUN

VAN DER WAALS FORCE BETWEEN QUARTZ PLATES AS A
FUNCTION OF SEPARATION, D

D IN ANGST	F IN NT/SQ CM
10	274.378
20	17.1486
30	3.38738
40	1.07179
50	.439005
60	.211711
70	.114277
80	6.69868E-02
90	4.18196E-02
100	2.74378E-02
110	1.87404E-02
120	1.32320E-02
130	9.60674E-03
140	7.14228E-03
150	5.41982E-03
160	4.18668E-03
170	3.28514E-03
180	2.61372E-03
190	2.10540E-03
200	1.71486E-03
300	3.38739E-04
400	1.07179E-04
500	4.39005E-05
600	2.11712E-05
700	1.14277E-05
800	6.69868E-06
900	4.18195E-06
1000	2.74378E-06
1100	1.87404E-06
1200	1.32320E-06
1300	9.60674E-07
1400	7.14228E-07
1500	5.41981E-07
1600	4.18668E-07
1700	3.28514E-07
1800	2.61372E-07
1900	2.10540E-07
2000	1.71486E-07
2100	1.41082E-07
2200	1.17127E-07
2300	9.80474E-08
2400	8.26998E-08
2500	7.02408E-08
2600	6.00421E-08
2700	5.16291E-08
2800	4.46393E-08
2900	3.87933E-08
3000	3.38738E-08
3100	2.97100E-08
3200	2.61667E-08

PAGE 2

D IN ANGST	F IN NT/SQ CM
3300	2.31363E-08
3400	2.05321E-08
3500	1.82842E-08
3600	1.63358E-08
3700	1.46400E-08
3800	1.31588E-08
3900	1.18602E-08
4000	1.07179E-08
4100	9.70988E-09
4200	8.81763E-09
4300	8.02556E-09
4400	7.32046E-09
4500	6.69113E-09
4600	6.12799E-09
4700	5.62287E-09
4800	5.16874E-09
4900	4.75954E-09
5000	4.39005E-09
5100	4.05573E-09
5200	3.75263E-09
5300	3.47733E-09
5400	3.22682E-09
5500	2.99846E-09
5600	2.78995E-09
5700	2.59926E-09
5800	2.42458E-09
5900	2.26434E-09
6000	2.11711E-09
6100	1.98166E-09
6200	1.85688E-09
6300	1.74176E-09
6400	1.63542E-09
6500	1.53708E-09
6600	1.44602E-09
6700	1.36160E-09
6800	1.28326E-09
6900	1.21047E-09
7000	1.14277E-09
7100	1.07973E-09
7200	1.02098E-09
7300	9.66179E-10
7400	9.15003E-10
7500	8.67170E-10
7600	8.22423E-10
7700	7.80524E-10
7800	7.41261E-10
7900	7.04435E-10
8000	6.69868E-10
8100	6.37396E-10
8200	6.06867E-10

PAGE 3

D IN ANGST	F IN NT/SQ CM
8300	5.78145E-10
8400	5.51102E-10
8500	5.25622E-10
8600	5.01597E-10
8700	4.78930E-10
8800	4.57529E-10
8900	4.37310E-10
9000	4.18195E-10
9100	4.00114E-10
9200	3.82999E-10
9300	3.66790E-10
9400	3.51429E-10
9500	3.36864E-10
9600	3.23046E-10
9700	3.09929E-10
9800	2.97471E-10
9900	2.85633E-10
10000	2.74378E-10

READY

TABLE A II

RJ-UN
 VAN DER WAALS FORCE BETWEEN QUARTZ PLATES AS A
 FUNCTION OF SEPARATION, D
 LONG RANGE WITH WATER FILM

D IN ANGST	F IN NT/SQ CM
10	1186.1
20	74.1313
30	14.6432
40	4.63321
50	1.89776
60	.915202
70	.494003
80	.289576
90	.180781
100	.11861
110	8.10123E-02
120	5.72001E-02
130	4.15287E-02
140	3.08752E-02
150	2.34292E-02
160	1.80985E-02
170	1.42012E-02
180	1.12988E-02
190	9.10138E-03
200	7.41313E-03
300	1.46432E-03
400	4.63321E-04
500	1.89776E-04
600	9.15202E-05
700	4.94003E-05
800	2.89576E-05
900	1.80780E-05
1000	1.18610E-05
1100	8.10123E-06
1200	5.72001E-06
1300	4.15287E-06
1400	3.08752E-06
1500	2.34291E-06
1600	1.80985E-06
1700	1.42012E-06
1800	1.12989E-06
1900	9.10138E-07
2000	7.41313E-07
2100	6.09880E-07
2200	5.06327E-07
2300	4.23848E-07
2400	3.57501E-07
2500	3.03642E-07
2600	2.59554E-07
2700	2.23186E-07
2800	1.92970E-07
2900	1.67699E-07
3000	1.46432E-07
3100	1.28433E-07
3200	1.13115E-07

P IN ANGST	F IN NT/SQ CM
3300	1.00015E-07
3400	8.87577E-08
3500	7.90404E-08
3600	7.06174E-08
3700	6.32871E-08
3800	5.68836E-08
3900	5.12700E-08
4000	4.63321E-08
4100	4.19746E-08
4200	3.81175E-08
4300	3.46935E-08
4400	3.16454E-08
4500	2.89249E-08
4600	2.64905E-08
4700	2.43069E-08
4800	2.23438E-08
4900	2.05749E-08
5000	1.89776E-08
5100	1.75324E-08
5200	1.62221E-08
5300	1.50321E-08
5400	1.39491E-08
5500	1.29620E-08
5600	1.20606E-08
5700	1.12363E-08
5800	1.04812E-08
5900	9.78844E-09
6000	9.15201E-09
6100	8.56648E-09
6200	8.02703E-09
6300	7.52939E-09
6400	7.06971E-09
6500	6.64459E-09
6600	6.25095E-09
6700	5.88603E-09
6800	5.54736E-09
6900	5.23269E-09
7000	4.94003E-09
7100	4.66754E-09
7200	4.41359E-09
7300	4.17667E-09
7400	3.95584E-09
7500	3.74867E-09
7600	3.55523E-09
7700	3.37411E-09
7800	3.20437E-09
7900	3.04518E-09
8000	2.89575E-09
8100	2.75538E-09
8200	2.62351E-09

REPRODUCIBILITY OF THIS
ORIGINAL PAGE IS 100%

PAGE 3

D IN ANGST F IN NT/SQ CM

8300	2.49925E-09
8400	2.38234E-09
8500	2.27220E-09
8600	2.16834E-09
8700	2.07035E-09
8800	1.97784E-09
8900	1.89044E-09
9000	1.80781E-09
9100	1.72964E-09
9200	1.65566E-09
9300	1.58559E-09
9400	1.51918E-09
9500	1.45622E-09
9600	1.39649E-09
9700	1.33978E-09
9800	1.28593E-09
9900	1.23476E-09
10000	1.18610E-09

READY

APPENDIX III. GRAPH OF VAN DER WAALS FORCE BETWEEN PLATES AS A FUNCTION
OF DISTANCE.

