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for
GRANT NAGW-533

LABORATORY EVALUATION AND APPLICATION OF
MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED
CONDITIONS FOR PLANETARY ATMOSPHERES

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I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO$_2$ was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, results obtained for the microwave opacity from gaseous H$_2$SO$_4$ under simulated Venus conditions, during the first two years of Grant NAGW-533 (February 1, 1984 through January 31, 1986), showed that not only was the opacity from H$_2$SO$_4$ much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986a). The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has
been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

In the first two years of Grant NAGW-533 (i.e., February 1, 1984 through January 31, 1986), this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid (H$_2$SO$_4$) under Venus atmospheric conditions. The initial results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Measurements of the microwave properties of the vapors accompanying liquid H$_2$SO$_4$ also resulted in more accurate estimates of the vapor pressure behavior of sulfuric acid, which are critical for modeling the behavior and structure of the Venus atmosphere. Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous H$_2$SO$_4$ at wavelengths from 1.2 to 22 cm under simulated Venus conditions. Additional measurements of the vapor pressure behavior of sulfuric acid were also made. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of H$_2$SO$_4$ and SO$_2$ abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort have been especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous H$_2$SO$_4$ in these wavelength ranges has finally explained what were thought to be inconsistencies between measurements of the absorption
in the Venus atmosphere at 13.3 and 3.6 cm wavelengths and those obtained in
the 1 to 3 cm wavelength range. We describe these effects, and the resulting
limitations they place on abundances of gaseous $H_2SO_4$ and $SO_2$ in the Venus
atmosphere, in a paper entitled, "Evaluation of the Microwave Spectrum of
Venus in the 1.2 to 22 Centimeter Wavelength Range Based on Laboratory
Measurements of Constituent Gas Opacities," which was submitted in February
(1986) and has been published in The Astrophysical Journal (v. 310, pp. 404-
414, November 1, 1986--Galley Proofs included as Appendix 6).

Additional activities conducted during this grant year (February 1, 1986
through January 31, 1987) involving application of our laboratory measurements
of simulated Venus atmospheres have included assistance to the Magellan
program office (J.P.L.) in characterizing effects of the atmosphere on planned
microwave radar and radiometric experiments originally planned to characterize
surface and geologic parameters. Our laboratory measurements have also
suggested that a substantial dip in the Venus microwave emission, related to
the abundance of gaseous sulfuric acid, should exist near the 2.25 cm
wavelength. Since no observations of the Venus emission at this wavelength
have ever been published, we have received permission to use the 140-foot NRAO
telescope in May 1987 to not only confirm the presence of the predicted dip,
but to use such a dip to determine a planet-wide average for sulfuric acid
vapor abundance below the main cloud layer. (Support for travel to NRAO and
use of the telescope will be provided by NRAO in response to our proposal,
"Observations of the Venus Microwave Spectrum at 1.9 and 2.25 cm" - attached
as Appendix 5.)

The highest priority activity for this grant year has been laboratory
measurements of the microwave properties of the simulated atmospheres of the
outer planets and their satellites. As described in a previous Annual Status Report for Grant NAGW-533 (February 1, 1985 through January 31, 1986), our planetary atmospheres simulator underwent a major redesign late last year to permit measurements of the microwave properties of simulated Jovian atmospheres. Further developmental work has been necessary in order to minimize the safety risks involved with high pressure simulations using hydrogen gas at very low temperatures, as required for the outer planets. A more complete description of this work is given in Section II of this report. However, since many of the low temperature, high pressure tests were initially conducted using the nonexplosive gas nitrogen (N₂), which is the primary constituent of the atmosphere of the Saturnian satellite Titan, useful data for microwave refractivity and absorptivity of nitrogen atmospheres under simulated Titan conditions has been obtained at frequencies from 2.2 GHz to 21.7 GHz (1.3 cm to 13.7 cm). Such data is useful in interpretation of both Voyager 1 radio occultation measurements of the Titan atmosphere, as well as radio astronomical observations of Titan.

The most significant accomplishment of this grant year has been the successful completion of laboratory measurements of the microwave opacity of gaseous ammonia (NH₃) in a hydrogen/helium (H₂/He) atmosphere, under simulated conditions for the outer planets. These measurements were conducted at frequencies from 1.3 GHz to 22 GHz (wavelengths from 1.3 cm to 22 cm), at temperatures from 178 K to 300 K, and under total pressures reaching as high as 6 atmospheres. Such measurements have long been sought by a number of researchers working on inferring ammonia abundance profiles in Jovian atmospheres. (See, for example, de Pater and Massie, 1985, or West et al., 1986.) Our measurements represent the first time that measurements of the
microwave absorption of gaseous ammonia under simulated conditions for the outer planets have been conducted. The results of these measurements, and their effect on the interpretation of microwave opacity data obtained both from Voyager radio occultation measurements made at 13 cm and 3.6 cm wavelengths, and from radio astronomical observations in the 1.3 cm to 22 cm wavelength range, are discussed in Section IV of this report.

Beyond this current grant year, our goals are to continue such laboratory measurements of the microwave absorption and refraction from other potential microwave absorbers contained in the outer planets' atmospheres, including methane (CH₄), water vapor (H₂O), and phosphine (PH₃), as well as additional high-sensitivity measurements of the absorption from gaseous NH₃ at 13.3 and 18.5 cm. We likewise would hope to be able to pursue a program of further analysis and application of these results to microwave data for the outer planets, such as Voyager Radio Occultation experiments and earth-based radio astronomical observations.

Of equal importance, we feel however, would be the further analysis and application of our laboratory results for the microwave absorption from gaseous H₂SO₄ in the Venus atmosphere. Our long term goal would be a detailed analysis of available multi-spectral microwave opacity data from Venus, including data from the Pioneer-Venus Radio Occultation experiments and earth-based radio and radar astronomical observations, such as the kinds which have been performed at the NRAO Very Large Array (VLA) and at stations in the Deep Space Network (DSN). The new measurements of Venus microwave emission at 2.25 cm and 1.9 cm made with the NRAO 140-foot telescope will be an especially important contribution to this data set. This would provide a chance to determine both spatial and temporal variations in the abundances of both H₂SO₄ and SO₂ in the Venus atmosphere.
Further discussion of proposed future activity, both regarding the outer planets and Venus, is included in the accompanying proposal to NASA entitled, "Laboratory Evaluation and Application of the Microwave Properties of Simulated Planetary Atmospheres."

II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the first two Annual Status Report(s) for Grant NAGW-533 (February 1, 1984 through January 31, 1985 and February 1, 1985 through January 31, 1986). It is also discussed at length in Steffes (1985 and 1986b). The updated simulator system, shown in Figure 1, is currently configured for simulations of the outer planets. Measurements of the microwave opacity and refractivity of test gas mixtures can be performed at frequencies from 1.3 GHz to 27 GHz (wavelengths from 1.2 cm to 22 cm). While the pressure chamber itself is capable of containing pressures up to 10 atmospheres, and the temperature chamber is capable of achieving temperatures as low as 150 K, it has been found that the combination of high pressures and low temperatures can create a substantial problem with sealing the pressure chamber. In previous simulations, small leakages from the pressure vessel presented little or no danger to the experimenters. The use of gaseous hydrogen (H₂) in the outer planets simulations has required the development of new procedures and equipment for conducting such simulations. Such precautions have included the addition of a hydrogen leakage sensor which is placed inside the temperature chamber immediately outside the pressure vessel (see Figure 1). This sensor can
detect the potentially dangerous build-up of hydrogen gas within the freezer unit. A ventilation pump has also been provided which can be used to draw any escaping hydrogen gas out of the freezer compartment. Additional precautions have included the construction of ramps which allow all of the equipment to be moved out-of-doors to a concrete slab immediately adjacent to the laboratory. All experiments which employ gaseous hydrogen can thus be conducted out-of-doors in order to avoid any build-up of hydrogen gas within the laboratory. A covered outdoor storage area for hydrogen and helium gases has also been constructed in order to allow safe storage of these gases, and to expedite the out-of-doors experiments. It is noteworthy that funding for the hydrogen leakage sensor, for construction of the outdoor gas storage and experimental areas, and for special tools required for sealing the pressure vessel (over $2,000) has been provided by the Georgia Institute of Technology, in support of planetary atmospheres research at Georgia Tech. In addition, two vacuum sensors have been added to the system. These sensors not only allow accurate determination of pressure vessel evacuation, but they can also be used for accurately determining the abundances of microwave-absorbing test gases, which are typically very small at low temperatures, due to low saturation vapor pressures.

Initial testing of the new simulator configuration for the outer planets was conducted using nitrogen (N₂) gas both because of its relative safety as compared with hydrogen, and because measurements of both the refractivity and absorptivity of nitrogen under simulated conditions for the Saturnian satellite Titan were considered important for interpreting results of the Voyager 1 radio occultation studies of the Titan atmosphere (Lindal et al., 1983 and 1985).
The first tests of the system showed that the pressure vessel could easily hold pressures of 7 atmospheres at room temperature with essentially zero leakage. However, when the system was cooled to 150 K, significant leakage occurred. A quick review of pressure vessel construction can shed some light on this problem. The pressure vessel is essentially a 13-inch diameter stainless steel cylinder (0.375-inch wall thickness) with a 0.375-inch thickness stainless steel plate welded to the bottom of the cylinder. At the top of the cylinder is a 0.5-inch thick cylindrical flange which is welded to the cylinder. The top plate (0.5-inch thickness stainless steel) bolts to the flange using 16 stainless steel bolts, along with a pressure sealant O-ring, and sealant putty which is placed outside the O-ring. (See Figure 2.) All electrical connections are made via hermetically sealed feed-through connectors mounted on the top plate.

Two causes for the increased leakage rate at extremely low temperatures have been found. The first was simply due to metal contraction. When the pressure vessel was cooled to 150 K, it was found that most of the fastener bolts had loosened due to contraction of the stainless steel. Thus, removal of the cooled pressure vessel from the freezer was necessary so that the bolts could be tightened while it was still very cold, and it was then immediately replaced inside the freezer. The special lifting frame and pulley system which was designed for easily moving the pressure vessel in and out of the freezer has been especially useful for this task. However, even after retightening of the fastener bolts on the cold pressure vessel, substantial leakage still occurred.

The cause of the continued leakage was found to be the sealing materials used in the pressure vessel. As shown in Figure 2, the seal used when placing
the top plate on the pressure vessel consists of a 0.385-inch cross-section O-ring accompanied by a ring of putty-type sealing material. As was the case in the recent Space Shuttle accident, seals which normally function well at room temperature can malfunction when placed under extremely cold conditions. This is due to increasing brittleness of the O-ring and putty seals, accompanied by a higher probability of cracks developing due to the brittleness. Initially, as suggested by several seal material vendors, we used an O-ring made from vulcanized rubber and RTV-silicone putty. The relatively poor performance of this seal caused us to experiment with several other types of sealing materials. We found that nearly all materials typically used in such seals became brittle at 150 K. However, we found that O-rings fabricated from the material Viton were less likely to develop cracks at this temperature. It was also found that RTV-silicone became extremely brittle at 150 K, and was very susceptible to cracking. As a result, we now use a more fibrous silicone composite material for the sealing putty.

The overall result has been a pressure vessel capable of maintaining 6 atmospheres of pressure at a temperature of 150 K, with an acceptably small leak ratio. While the range of pressures which can be tested is not quite as large as originally hoped, the resulting range of temperatures and pressures does represent the range over which nearly all of the microwave opacity in the Jupiter atmosphere has been observed, and thus is extremely useful in interpretation of microwave opacity data from Voyager I and II radio occultation experiments, as well as from earth based radio astronomical observations, and, in the future, opacity measurements to be made using the Galileo probe. Likewise, the pressure-temperature ranges measured will be close enough to those over which microwave absorption or refraction has been measured in the
atmospheres of Saturn, Titan, Uranus, and Neptune, so that accurate estimates of abundances of microwave-absorbing constituents in these atmospheres can also be made.

III. EXPERIMENTAL APPROACH

The two key measurements made in the past grant year with the Georgia Tech planetary atmospheres simulator have been the measurements of the refractivity of gaseous nitrogen (N₂) under conditions similar to the Titan atmosphere and the measurements of the microwave absorption from gaseous ammonia (NH₃) under simulated conditions for the outer planets. It should be noted, however, that the techniques described for measuring refraction and absorption can be applied to any gases which are placed in the simulator.

The refractive index, \( n \), of a gas is the ratio of the velocity of an electromagnetic wave in a vacuum, \( c \), to the velocity of the wave in the presence of the gas, \( V_g \). Since the resonant frequencies of a microwave resonator are directly proportional to the velocity of electromagnetic waves within the resonator, the refractive index of a gas can be determined by comparing the frequency of a particular resonance when a vacuum is present within the resonator, \( f_v \), to the resonant frequency when the gas is present, \( f_g \). That is,

\[
\frac{c}{V} = \frac{f_v}{f_g}
\]

Refractivity, \( N \), is defined as being equal to \((n-1) \times 10^6\). Thus, it can be determined simply by measuring the change in center frequency of a given resonance after the gas has been introduced. That is,
\[ N = (n-1) \times 10^6 = \left[ \left( f_v - f_g \right) \right] f_g \times 10^6 \] 

Since the refractivity of a gas is directly proportional to the molecular density of the gas, \( \rho \), the refractivity is often expressed in a form which is normalized by molecular density in terms of the temperature and pressure of the gas. That is, \( \rho = \frac{P}{RT} \) where \( \rho \) is density in molecules per \( \text{cm}^3 \), \( P \) is the pressure in atmosphere, \( R \) is the ideal gas constant \( (1.362344 \times 10^{-22} \text{ cm}^{-3} \text{-atm/mol/K}) \), and \( T \) is the temperature in Kelvins. Thus, the density-normalized refractivity, \( N/\rho \), can be expressed as \( NRT/P \). It is often assumed that the density-normalized refractivity for a gas is independent of pressure, temperature, or frequency.

It is this assumption which allowed Lindal et al. (1983) to infer a temperature-pressure profile for Titan's atmosphere from measurements of atmospheric refractivity at 2.3 GHz (13 cm) made with Voyager 1. That is, by assuming an exponential atmosphere with surface temperature 94 K, and by assuming a constant value for the density-normalized refractivity of \( N_2 \) (the predominant constituent) to be \( 1.093 \times 10^{-17} \) N-units/molecule/cm\(^3\), the measured refractivity profile for Titan's atmosphere was used to infer a temperature-pressure profile for the atmosphere. This value for normalized refractivity was obtained by Essen and Froome (1951) based on a single measurement of the refractivity of nitrogen \( (N_2) \) under standard laboratory conditions at a single frequency (24 GHz). The need to confirm by laboratory measurement the nature of the refractive properties of nitrogen at 2.3 GHz under conditions similar to those of Titan is considered critical for the proper interpretation of Titan refractivity data.
In order to measure the refractivity of nitrogen, the pressure vessel and its microwave resonators (see Figure 1) must first be cooled to the desired temperature. Because of the large mass of the pressure vessel and microwave resonators (over 400 pounds), nearly 24 hours are required to reach thermal equilibrium at temperatures below 195 K using our current freezer system. Temperatures within the temperature chamber (freezer) are monitored by two temperature sensors: the first being placed within the chamber, but outside the pressure vessel; and the second being placed within the pressure vessel. Another technique used in monitoring the cooling of the resonator within the pressure vessel is to monitor its resonant frequencies. As the resonator is cooled, the resonant frequencies drop due to thermal contraction. Thus, when thermal stability is reached, the resonant frequencies likewise stabilize. After thermal stability is reached, a vacuum is drawn in the pressure vessel containing the two microwave resonators, and the center frequencies of the resonances of interest are then measured. For this experiment (refractivity of N\textsubscript{2}), resonances at 2.26 GHz (13.3 cm), 8.53 GHz (3.52 cm), 13.3 GHz (2.26 cm), and 21.77 GHz (1.38 cm) were used. A valve is then opened which admits the test gas (N\textsubscript{2} in this case) to the chamber. The nitrogen is admitted at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. As the gas is added, the shifting of the center frequencies of the various resonances can be observed. Once the desired pressure is reached (pressures between 1 atm and 6 atm were used in our experiments), the total frequency shift can be used to compute the refractivity, N, of the gas under those conditions. It should be noted that while the major source of uncertainty for our refractivity measurements is the frequency measuring capability of our system, two other sources of uncertainty
affect the accuracy of our determination of density-normalized refractivity, and those are our abilities to measure pressure and temperature. For example, in some early measurements a less accurate pressure gauge was used. Thus, the error bars for the measurement of density-normalized refractivity were larger than those for similar measurements made later. The results of these measurements are discussed in Section IV of this report.

The approach used to measure the microwave absorptivity of gaseous NH₃ in an H₂/He atmosphere is similar to that used previously by Steffes (1985 and 1986a) for simulated Venus atmospheres. As can be seen in Figure 1, the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of two cavity resonators at particular resonances from 1.34 GHz to 21.8 GHz. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

\[
\alpha = \left( Q_{L}^{-1} - Q_{C}^{-1} \right) \pi / \lambda
\]

where \( \alpha \) is absorptivity of the gas mixture in Nepers km\(^{-1}\). (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km\(^{-1}\) = 2 optical depths per km (or km\(^{-1}\)) = 8.686 dB km\(^{-1}\), where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a
possible factor-of-two ambiguity in meaning.) $Q_L$ is the quality factor of the cavity resonator when the gas mixture is present, $Q_C$ is the quality factor of the cavity resonator in a vacuum, and $\lambda$ is the wavelength (in km) of the test signal in the gas mixture.

In order to obtain a gas mixture with a sufficient amount of gaseous NH$_3$ so that microwave absorption is detectable using our system, temperatures at or above 170 K must be used. (This limit is set by the saturation vapor pressures for ammonia and by the sensitivity of our measurement system.) While this covers most of the temperature range in the Jupiter atmosphere over which radio occultation and radio astronomical experiments have detected microwave opacity (140-300 K), it is somewhat above the temperature range over which microwave opacity has been detected at Saturn. However, the measured temperature dependencies can be used to extrapolate to those temperatures. In order to conduct the required measurements, the pressure vessel and its microwave resonators must first be cooled to the desired temperature. As with the refractivity measurements, this requires over 24 hours of cooling in the temperature chamber. Before the cooling process begins, the chamber is filled with 6 atm of a neutral gas (either helium or nitrogen). This is because the pressure vessel achieves a much better seal if it is cooled while under pressure.

After thermal stability is reached, which can be monitored using both the temperature sensors and the resonant frequencies of the system, a vacuum is drawn in the pressure vessel containing the resonators, and the bandwidth and center frequency of each of resonances is then measured. For this experiment (absorption from NH$_3$), resonances at 1.34 GHz (22.3 cm), 2.25 GHz (13.3 cm), 8.53 GHz (3.52 cm), 13.3 GHz (2.26 cm), and 21.7 GHz (1.38 cm) were used.
A valve is then opened which allows the ammonia gas to enter the chamber. For the experiments conducted at 178 K, only about 2 torr NH\textsubscript{3} pressure was admitted so as not to cause saturation or condensation. For all other experiments (193 K and above), 20 torr NH\textsubscript{3} pressure was used. Measurements of the gaseous NH\textsubscript{3} pressure were made with the high accuracy thermocouple vacuum gauge tubes which are shown in Figure 1. Next, 5.4 atm of hydrogen (H\textsubscript{2}) and 0.6 atm of helium (He) are added. These gases are admitted to the chamber at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. The result is an atmosphere with 6 atm total pressure composed of 90 percent hydrogen, 10 percent helium, and approximately 4300 ppm ammonia (except for the measurements conducted at 178 K, where the ammonia abundance was approximately 430 ppm). The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure is then reduced by venting to 4 atm, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. The measured changes of bandwidth (Q) can then be used to compute the absorptivity of the gas mixture under the various pressure conditions.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any
derived pressure dependence is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency dependence of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Measurements made of the microwave refraction (and absorption) from Nitrogen ($N_2$) under simulated conditions for Titan are shown in Table I. While no absorption was measured at any frequency from 2.2 GHz to 21.7 GHz, upper limits for the opacity from $N_2$ can be set. This is especially important since $N_2$ may very well be the source of the 3.6 cm (8.4 GHz) opacity detected by Voyager I radio occultation studies of the Titan atmosphere (Lindal et al., 1985). Also shown in Table I is the refractivity, $N$, and the refractivity normalized by molecule number density. Since the normalized refractivity is used directly to determine atmospheric pressure from measurements of the atmospheric refractivity at a given altitude, an accurate expression for normalized refractivity at the temperature of that altitude is required.

When inverting the Voyager I refractivity data obtained at 2.3 GHz at Titan, in order to develop a temperature-pressure profile for Titan's atmosphere, Lindal et al. (1983) assumed an exponential atmosphere with surface temperature 94 K, and a constant value for the density normalized refractivity of gaseous $N_2$ ($1.093 \pm 0.0004$ N-units/molecule/cm$^3$). This value for normalized refractivity was obtained by Essen and Froome (1951) based on a
single measurement of the refractivity of nitrogen under standard laboratory conditions at a single frequency (24 GHz). This value is included, for comparison, in Table I. It should be noted that 4 additional data points are presented in this table, beyond those previously presented in Semiannual Status Report #5 (July 1986) for this grant. We have measured the refractivity of nitrogen over a wide range of temperatures (down to 156 K) and over a wide range of frequencies (2.2 GHz to 21.7 GHz) in order to determine whether any temperature or frequency variations of the density-normalized refractivity existed, and to estimate such effects on the interpretation of the refractivity data. While some variations were found between the measurements of the density-normalized refractivity of nitrogen, all were within experimental error of the values determined by Essen and Froome (1951) at 24 GHz and by Birnbaum et al. (1951) at 9.58 GHz. These results confirm that the density-normalized refractivity of nitrogen remains essentially constant, even at the very cold temperatures characteristic of the outer planetary systems. While our measurements did not go as low as the 94 K surface temperature of Titan, they do strongly indicate that the method for inferring the temperature-pressure profile of Titan's atmosphere from Voyager 1 refractivity data used by Lindal et al. (1983) was appropriate, and that the resulting profiles are accurate.

Measurements of the microwave absorption from NH$_3$ in a 90% hydrogen (H$_2$)/10% helium (He) atmosphere have been conducted over a wide range of temperatures, pressures, and frequencies. For measurements conducted at a temperature of 178 K, with an ammonia mixing ratio of only 430 ppm, the error bars are quite large as a percentage of the measured value, since the opacity of the gas mixture is small. For the measurements made at 193 K and 300 K,
where an ammonia mixing ratio of 4300 ppm was used, the error bars are much smaller. Data for absorption from the gas mixtures was obtained at all of five resonant frequencies previously mentioned: 1.34 GHz (22.3 cm), 2.25 GHz (13.3 cm), 8.53 GHz (3.52 cm), 13.3 GHz (2.26 cm), and 21.7 GHz (1.38 cm). However, due to the relatively low opacity exhibited by the gas mixtures at 1.34 GHz, and likewise due to the relatively poor sensitivity of our system at that lowest resonance, the data obtained at 1.34 GHz was fairly noisy, and will require further analysis and calibration. The complete results of all measurements at all other frequencies are presented in tabular form in Appendix 1, by frequency: 2.25 GHz (Table 1), 8.53 GHz (Table 2), 13.3 GHz (Table 3), and 21.7 GHz (Table 4). In addition to the measured values for absorption, and the accompanying error bars, the theoretically-derived values for the opacity for each gas mixture under the conditions of each measurement are also given for comparison.

A more useful presentation of this data is given in Figures 3, 4, and 5. In Figure 3, the measured values of the absorptivity are plotted as a function of frequency (wavelength), for total gas mixture pressures of 6, 4, and 2 atmospheres. All measurements shown in this figure were made at a temperature of 193 K and with an NH$_3$ mixing ratio of 4.3 x 10$^{-3}$. Also shown are plots of the theoretically-derived absorption spectra for such gas mixtures at these three pressures. These theoretically-derived NH$_3$ absorption spectra were taken from the calculations of De Pater and Massie (1985) and Berge and Gulkis (1976), both of which employ a modified Ben-Reuven line shape. Our theoretical spectra have also been corrected for the temperature, pressure, and mixing ratio conditions of our experiment. Figure 4 presents similar results for data taken at 300 K with an NH$_3$ mixing ratio of 4.3 x 10$^{-3}$. Figure 5
presents results for a temperature of 178 K and an NH$_3$ mixing ratio of only $4.3 \times 10^{-4}$, resulting in fairly large error bars, as previously discussed. It should be noted that the 13.3 GHz data points (2.26 cm) and data at 21.7 GHz taken on July 25, 1986, are not included in Figures 3, 4, and 5, both for the sake of clarity and because some calibration accuracy problems arose due to problems with connectors on that resonator, especially with the August 8 data set. Also, some data taken at 1 atm pressure was not included (even though it agrees well with the theoretical predictions for opacity at that pressure) for the sake of clarity.

Inspection of Figures 3, 4, and 5 shows that the laboratory data agree surprisingly well with the theoretical predictions for NH$_3$ opacity made using the modified Ben-Reuven lineshape. Our results provide what De Pater and Massie (1985) characterized as "information which is desperately needed," in that they provide answers to a number of questions which have been published regarding the NH$_3$ spectrum. The key finding is that the modified Ben-Reuven line shape appears to correctly describe the shape of the absorption spectrum from gaseous ammonia in a hydrogen/helium atmosphere in the 1.3 to 13.3 cm wavelength range even under the temperature-pressure conditions characteristic of the Jovian atmosphere. This finding answers some questions and raises others. For example, both de Pater and Massie (1985) and West et al. (1986) recognized that, based on interpretation of the Jovian emission spectrum in the 10 to 20 cm wavelength range using the theoretically-derived absorption spectrum from ammonia, opacity at pressure levels greater than 2 atm had to exceed the amount which would be caused by the solar abundance of ammonia. As a result, both sets of authors concluded that either the theoretical lineshape was incorrect and understated the opacity of ammonia at these wavelengths by a
factor of 1.5-2.0, or the ammonia abundance was greater than its solar abundance by a factor of 1.5-2.0 at pressures greater than 2 atm, or that an extra opacity source, possibly H$_2$O condensate, was present. Thus, since our measurements of the ammonia opacity at 13.3 cm (2.25 GHz) agree quite closely with the theoretically-derived values for opacity, and since our preliminary work at 22.3 cm (1.34 GHz) is likewise consistent with the theoretical lineshape, it would appear that either the ammonia abundance is greater than solar by a factor of 1.5-2.0 in the deeper layers of the Jovian atmosphere, or that an extra opacity source is present.

Either or both of these scenarios are possible. For example, based on the somewhat localized Voyager 1 radio occultation studies, Lindal et al. (1981) concluded that at a pressure of 1 atm, the ammonia mixing ratio was $2.2 \times 10^{-4}$ (1.5 times the solar abundance). On the other hand, de Pater and Massie (1985) found that in order to best match Jupiter's microwave emission spectrum (planet-wide average, 1.3-20 cm), ammonia abundances of $3 \times 10^{-5}$ for pressures less than 1 atm, $1.5 \times 10^{-4}$ at 1.5 atm, and $2.5 \times 10^{-4}$ at pressures greater than 2 atm were required. However, they did suggest that a solar abundance of ammonia ($1.5 \times 10^{-4}$) at pressures greater than 1.5 atm would be consistent with the observed emission spectrum if a water cloud with mass density 35 g/m$^3$ were present and if the Ben-Reuven lineshape used in their calculation was correct. West et al. (1986) agreed by stating the only direct evidence which suggest that an H$_2$O abundance great enough to form a cloud actually exists is the 10 to 20 cm spectrum, and only if it can be shown that NH$_3$ is incapable of supplying the required opacity. Our work is especially important in that it shows that a solar abundance of ammonia is indeed incapable of supplying the required opacity.
It may be possible to differentiate between the remaining two suggested possibilities (an ammonia abundance which is 1.5-2.0 times the solar abundance or a cloud layer in the 5-6 atm region) if a more accurate estimate of the frequency dependence of the atmosphere opacity in the 10 to 20 cm wavelength range could be obtained. We would then be able to determine which combinations of gaseous NH$_3$, liquid H$_2$O, and gaseous H$_2$O (in the 5 to 6 atm pressure range) could account for such a frequency (wavelength) dependence. In order to support this effort, we are proposing to make additional measurements in the laboratory of the microwave absorption from gaseous NH$_3$ and gaseous H$_2$O under simulated conditions for the 5 to 6 atm pressure range of the Jovian atmosphere. These measurements would be conducted at 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm), and would take advantage of special adjustments to the atmospheric simulator so as to maximize sensitivity in the 10 to 20 cm wavelength range. We would also hope to further study the possible contributions to the opacity from the cloud layer itself, or from other unrelated gases such as phosphine (PH$_3$). For further discussion of proposed future activity, see the attached proposal, "Laboratory Evaluation and Application of the Microwave Properties of Simulated Planetary Atmospheres."

Another important aspect of our measurements of the opacity from ammonia under simulated Jovian atmospheric conditions are the results obtained at 21.7 GHz (1.38 cm). These results (shown in Figures 3-5 and listed in Appendix 1, Table 4) indicate that the absorption from ammonia is great enough at the 1.3 cm wavelength so as to require its depletion at pressures less than 0.5 atm in the Jovian atmosphere, as suggested both by Klein and Gulkis (1978) and By de Pater and Massie (1985). This drop in NH$_3$ abundance at altitudes above the 0.5 atm pressure level was also inferred by Lindal et al. (1981).
from Voyager 1 measurements of 13 cm and 3.6 cm radio opacity. Our laboratory measurements of the NH$_3$ opacity at these wavelengths confirms this conclusion.

Although the temperatures over which most of our laboratory measurements have been made are somewhat higher than those at Saturn over which microwave opacity has been detected, our results can be extrapolated to allow analysis of the Saturn microwave emission spectrum and radio occultation data, similar to what we have done for Jupiter. In fact, a very similar problem regarding the presence of a potential water cloud, or an overabundance of NH$_3$ (by a factor of 3 compared to its solar abundance) in the deep Saturn atmosphere could be resolved by applying our current results and future planned measurements. (Reference: Attached proposal).

At present, we are conducting preliminary studies of the microwave refraction and absorption from methane CH$_4$. These results for refractivity at 13.3 cm and 3.52 cm will be especially important for correctly inferring the temperature-pressure profiles of the atmospheres of Titan and Uranus from Voyager refraction profiles near these wavelengths, since both have been shown to have significant quantities of methane. In addition, since the ammonia abundance at Uranus has been shown to be substantially depleted (see Gulkis et al., 1978), any measurement of, or limitation on, the microwave opacity from methane will be especially important for interpretation of any radio opacity data (both radio occultation measurements and radio astronomical observations). Likewise, due to the depleted ammonia abundance, the role of water vapor and condensed water as microwave absorbers in the deep Uranus atmosphere becomes more critical, hence, the further need for the proposed measurements of the microwave absorption from gaseous H$_2$O under simulated...
conditions for the deep atmospheres of the outer planets. The results for ammonia, methane, and water vapor will likewise be invaluable for the interpretation of data from Neptune.

V. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the grant year, a paper was completed and accepted for publication in The Astrophysical Journal, describing results and applications of experiments performed during the second year of Grant NAGW-533 (Steffes, 1986a). This paper is described at length in Section I of this report. In May 1985, a paper was presented at the Conference on Jovian Atmospheres, at the Goddard Institute for Space Studies (New York) entitled, "Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Simulated Conditions for the Outer Planets." This paper described our plans and capabilities for simulating outer planets atmospheres and measuring microwave properties of those atmospheres. At the beginning of this year, a revised manuscript was completed and submitted for inclusion in a NASA Conference Proceedings for this conference (Steffes, 1986b).

Our most recent results for the laboratory measurements of the microwave opacity of ammonia under Jovian conditions are being presented at the 18th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society (AAS/DPS Meeting - Abstract attached as Appendix 2). In a related meeting being held in conjunction with the DPS meeting, entitled, "Laboratory Measurements for Planetary Science," we are presenting a summary of our laboratory activities, and their application to the interpretation of planetary radio absorptivity and refractivity data. Support for travel to Paris for these meetings has been provided by the Georgia Tech Research
Corporation (GTRC) in support of planetary atmospheres research at Georgia Tech.

The results of our laboratory measurements of the microwave opacity of ammonia and their application to the interpretation of microwave opacity data from Jupiter are currently being prepared for submission as a paper to Icarus. The paper will be entitled, "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH3) under Simulated Conditions for Jovian Atmospheres," and will be submitted in December. Similarly, our measurements of the refractive and absorption properties of nitrogen under conditions similar to the Titan atmosphere are being prepared for publication. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman, regarding radio astronomical measurements of Venus opacity), at the Stanford Center for Radar Astronomy (V. Eshleman, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Michael J. Klein, Michael Janssen, and Samuel Gulkis regarding radio astronomical observations of the outer planets, and A. J. Kliore regarding the Pioneer-Venus Radioscience Program). We have also had discussions with members of the Magellan Program Office at JPL with regard to using results from our laboratory measurements of simulated Venus atmospheres in planning mission experiments.

Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech.

Several publications which describe our research activities in the Planetary Atmospheres to the public have also been released. Two examples are
shown in Appendix 3. The first, entitled "Probing Venus," was a two-page color article describing the results of our work simulating the microwave properties of the Venus atmosphere, and appeared in the publication Research Horizons, published by the Georgia Institute of Technology (Winter 1986, Volume 3, Number 4, pp. 8-9). We have also attached an article which appeared in the Tuesday, June 10 edition of The Atlanta Constitution (pg. 34a) and in the Tuesday, June 10 edition of The Atlanta Journal (pg. 22a), entitled "Tech Making Down-to-Earth Studies of Outer Space." This article likewise appeals to a large public audience. As in the past, we have maintained contact with members of the Georgia congressional delegation, both by telephone and by letter (see Appendix 4), keeping them aware of our work and aware of the need for continued support of the solar system exploration program.

VI. CONCLUSION

This grant year (February 1, 1986 through January 31, 1987) has been both exciting and productive. Two papers have been published (Astrophys. J and NASA-CP), two conference papers are being presented, and two new papers are being submitted for publication. After long, arduous work with our simulator, measurements of the refractivity and absorptivity of nitrogen under conditions similar to those for Titan were completed. The most significant measurements, however, were those of the microwave absorption from gaseous ammonia (NH₃) under simulated conditions for the Jovian atmospheres (in a 90% H₂/10% He atmosphere at temperatures from 178 K to 300 K and pressures from 1 to 6 atm) over wavelengths from 1.3 to 22 cm. The results of these measurements are critical in that they confirm the theoretical calculation of the ammonia opacity using the Ben-Reuven lineshape. The application of both these
results, and results obtained previously, to planetary observations at microwave frequencies has been especially rewarding.

Applications of our results for ammonia to radio astronomical observations of Jupiter in the 1.3 to 20 cm wavelength range show that a strong depletion of ammonia for pressures less than 0.5 atm in the Jovian atmosphere must exist. They also show that a solar abundance of ammonia gas cannot account for the opacity inferred from the 10 to 20 cm emission spectrum in the 5 to 6 atm pressure range of the Jovian atmosphere, suggesting either an over-abundance of NH$_3$ vapor, or the presence of another microwave absorber such as a dense cloud or gases such as H$_2$O which would accompany clouds at that altitude. We are proposing to further investigate the differences in the frequency dependence of opacity from gaseous H$_2$O and gaseous NH$_3$ in the laboratory under Jovian conditions so as to better determine which of the two sources of opacity are present and in what relative quantities. (See accompanying proposal.)

Application of our results for gaseous H$_2$SO$_4$ under simulated Venus conditions, obtained during the previous grant year, have shown that a 15 ppm abundance of gaseous sulfuric acid (on average) below the main cloud layer exists, and approximately 150 ppm of SO$_2$ exists in the same region. Of more importance, our results show that certain portions of the Venus emission spectrum (especially around 13 GHz) are especially strongly affected by the sulfuric acid vapor abundance. Since no observations of the Venus emission at this wavelength have ever been published, we will use newly acquired data from NRAO in order to confirm the gaseous H$_2$SO$_4$ abundance on a planet-wide basis. It is also important to recognize that due to instrumental limitations of the Venus atmospheric probes, no direct measurements of the abundance of gaseous
H$_2$SO$_4$ exist for the subcloud region of Venus. However, Steffes (1985) showed that the major source of the microwave absorptivity measured by the Pioneer-Venus Radio Occultation experiments is gaseous H$_2$SO$_4$. Since the current radio occultation "season" is expected to yield the best measurements since the initial Pioneer-Venus radio measurements, we are hoping to become involved directly with the interpretation of that data in order to develop localized profiles for gaseous H$_2$SO$_4$ abundance in the Venus atmosphere. This information would be of key importance in better modeling the Venus atmosphere.
VII. REFERENCES


VIII. KEY FIGURES
<table>
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<tr>
<th>Frequency, GHz</th>
<th>Conditions</th>
<th>Refractivity ( N = 10^6(n-1) )</th>
<th>Refractivity Normalized by Number Density ( (N\text{-units/molecule/cm}^3) )</th>
<th>Absorptivity ( \text{dB/km} )</th>
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<td>( (1.0886 \pm .104) \times 10^{-17} )</td>
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<td>( (1.099 \pm .088) \times 10^{-17} )</td>
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<td>( (1.0938 \pm .0004) \times 10^{-17} )</td>
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Figure 1: Block Diagram of updated Georgia Tech Planetary Atmospheres Simulator, as configured for measurements of microwave properties of gases under simulated conditions for the outer planets.
Figure 2: Sketch of pressure vessel, as viewed from above, with the top plate removed. Note the welded flange, with groove for the O-ring.
Figure 3: Measured microwave absorption from ammonia ($4.3 \times 10^{-3}$ by volume) in a 90% hydrogen/10% helium atmosphere as a function of frequency (wavelength) at 193 K. Also shown are the theoretically-derived absorption profiles.

Wavelength, cm.

Absorption, dB/Km

Frequency, GHz

- 6 atm. 8/13/86
- 4 atm. 8/13/86
- 2 atm. 8/13/86
- 6 atm. 7/25/86
- 4 atm. 7/25/86
- 2 atm. 7/25/86
Figure 4: Measured microwave absorption from ammonia \((4.3 \times 10^{-3}\ \text{by volume})\) in a 90\% hydrogen/10\% helium atmosphere as a function of frequency (wavelength) at 300 K. Also shown are the theoretically-derived absorption profiles.
Figure 5: Measured microwave absorption from ammonia (4.3 $\times 10^{-4}$ by volume) in a 90% hydrogen/10% helium atmosphere as a function of frequency (wavelength) at 178 K. Also shown are the theoretically-derived absorption profiles.

![Graph showing microwave absorption as a function of frequency and wavelength for ammonia in a 90% hydrogen/10% helium atmosphere at 178 K. The graph includes theoretically-derived absorption profiles and data points for 6 atm. and 3 atm. on 7/14/86.](image-url)
IX. APPENDICES
### Table 1: Absorption Summary for 2.25 GHz

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Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH₃) under Simulated Conditions for Jovian Atmospheres

P. G. Steffes, J. M. Jenkins, M. F. Selman, W. W. Gregory (Georgia Institute of Technology)

We have recently completed laboratory measurements of the microwave opacity of gaseous ammonia (NH₃) under simulated conditions for Jovian atmospheres (i.e., as a minor constituent in a 90% H₂/10% He atmosphere, with temperatures ranging from 170 K to 300 K, and total atmospheric pressures ranging from 1 to 6 atmospheres). While the shapes of the absorption spectra, observed at wavelengths from 1.3 cm to 22 cm, seem consistent with theoretically derived expressions for the microwave opacity from NH₃ obtained using the Ben-Reuven line shape, some small variations from the theoretical absorption spectra may exist. These measurements are important for the interpretation of radio absorptivity data obtained from both radio astronomical and radio occultation observations of the outer planets. They are especially important for use in determining the abundance and distribution of NH₃ in the atmosphere of Jupiter.
PROBING VENUS

Remote sensing technology gives researchers detailed new data about the Venusian atmosphere

Remote sensing from space has given the human race a new perspective on itself. With vivid satellite portraits of the earth, planners can see pollution in coastal marshlands, insect infestation, fishing beds and a wealth of other important information. Now, the same basic technology is letting scientists map the atmospheres of planets millions of miles away. At Georgia Tech, Dr. Paul Steffes, an assistant professor of electrical engineering, has used remote sensing to "see" the make-up and structure of sulfuric acid in the mid-atmosphere of Venus.

The raw data for his research were microwave signals transmitted by Mariner and PVO spacecraft as they flew behind Venus. Before returning to earth, these transmissions penetrated the Venusian atmosphere, where they were either bent (refracted) or wandering (reflected). The resulting signal modifications were collected by the Deep Space Network of antennas at stations in California, Spain and Australia.

"Most of the refraction is due to the density of the atmosphere," says Steffes. "It's like a light wave being bent as it passes through the ocean."

Weaker signals indicate an absorption of electromagnetic energy by the atmosphere. Since only certain gases absorb electromagnetic waves, this information becomes a key for accomplishing this on a parts-

per-million level through a laboratory simulation of Venus' mid-atmosphere. With funding from NASA, he and his students built a small pressure chamber. They first injected gases known to be in the planet's atmosphere and subjected them to pressures and temperatures which exist at certain altitudes above Venus. Next, they propagated signals through the chamber at the same frequencies originally transmitted by NASA spacecraft back to earth. Finally, they manipulated the concentrations of gases present until they were able to bring about the same degree of signal attenuation or refraction observed through remote sensing. Through these experiments, Steffes' group found that they could calculate concentrations of a microwave-absorbing gas, sulfuric acid, in the mid-atmosphere of Venus.

The accuracy of this method has proven greater than achieved by probes dropped into the Venusian atmosphere by American spacecraft earlier exploring the planet. Simulations using remote sensing data have revealed sulfuric acid vapor abundances down to an order of 10 parts-per-million. The perfected (computerized) these experiments has been somewhat surprising. "Approximately 20 parts-per-million of sulfuric acid vapor exist below the clouds of Venus," Steffes says. "In the altitude range of 30 to 48 kilometers there is sulfuric acid vapor which condenses at 48 kilometers. Initially, it wasn't thought that this amount of sulfuric acid might have existed. It was thought that sulfuric acid clouds cannot be formed directly below the clouds only sulfur dioxide existed." Steffes' current work growing out of remote sensing experiments he made in the early 1980s as a doctoral degree student at Stanford University. However, his interest in the field began during his undergraduate years at the Massachusetts Institute of Technology. There, remote sensing techniques were being used to learn about the abundance of certain gases in the upper parts of the earth's atmosphere. "At MIT, we were able to infer how much oxygen and water vapor was in the earth's atmosphere," Steffes says, "so we said if it works for us here we can apply it to other planets too."

The implications of his findings for Venus are uncertain, but Steffes says that they could provide evidence for volcanic activity on the planet. He also believes that knowledge of how sulfuric acid moves in the atmosphere of Venus possibly can help scientists better understand the acid rain problem on earth.

With the Venus simulations completed, NASA is sponsoring Steffes in a program to improve his remote sensing data on outer planets. Voyagers 1 and 2 have already passed Jupiter and Saturn. Last January, Voyager 2 started sending data back from Uranus. In another three years, the ship will make its approach to Neptune and its moon Triton.

To simulate the atmosphere of these planets, Steffes has acquired a deep freeze which can reach -120° C., well above the -190° C. atmospheric temperature of Uranus but close enough for accurate extrapolations of data.

The outer planets will present an entirely different challenge than Venus. According to Steffes, some scientists are convinced that "that never quite made it" - that is, large gaseous balls with relatively small rocky cores. Their atmospheres are very highly pressurized and composed primarily of hydrogen and helium, gases which do not absorb microwave radiation well.

In his experiments, Steffes will fill the deep freeze with these gases, then start adding other gases, such as ammonia and methane, trying to match the attenuation and refraction of the Voyager microwave data. The work on Jupiter, Saturn and Uranus will probably take a year and a half.

What will be the value of his findings? "We'll be learning a lot about the nature of the formation of planets," Steffes says. "That tells us in the long run about how the universe is formed and whether there may be planets around other stars. So the applications are long term, obviously. They aren't necessarily something where we're going to be able to turn it into a new widget next week, but the implications are pretty significant."

ORIGINAL PAGE IS OF POOR QUALITY

Appendix 3.
Tech making down-to-Earth studies of outer space

By Robert Lee Holtz
Science/Medicine Writer

Inside a squat, square freezer at the Georgia Institute of Technology, scientists are seeking clues to the frigid gases that envelop the solar system's outer planets in a project that may yield new insights into the evolution of Earth's atmosphere.

By interpreting subtle distortions in radio signals from the Voyager 2 space probe, Georgia Tech researchers are mapping the exotic atmospheres of Jupiter, Saturn, Uranus and Titan, Saturn's largest moon, then duplicating the volatile blend of gases in a pressurized laboratory freezer.

Analyzing microwave signals from unmanned Mariner and Pioneer probes, they have detected corrosive clouds of sulfuric acid that dot the skies of Venus. Using large Earth-based radiotelescopes, they hope one day to track the daily weather on Venus or detect any active volcanoes.

"We are doing this research not only so we could better understand these other planets, but also to understand how our own atmosphere might be evolving," says Dr. Paul Steffes, a Georgia Tech professor of electrical engineering who leads the research team.

"We are learning how fragile our own atmospheric balance is — because if the scales tip one way, we could end up very hot like Venus or, if the scales tip the other way, we could end up like Mars. It is a very delicate balance that keeps our atmosphere so amenable to life."

None of the probes carries instruments designed to detect atmospheric gases. But when a satellite passes behind a planet or moon, the radio signals being transmitted back to Earth are garbled briefly by interference from the gases surrounding the other planet. Steffes has turned that distortion into a tool for mapping the alien atmosphere itself.

"Using the U.S. Deep Space Network, we are able to measure the amount of weakening in the microwave signal as it passed through the planet's atmosphere," Steffes says. "The amount of microwave radiation absorbed or refracted depends on what gases are in the planet's atmosphere. The question for us is what gases and how much," he says.

Any weakening of the satellite's signal indicates the presence of chemical compounds that absorb microwave energy. By pinpointing the frequency blocked, researchers can decipher the chemical signature of gases such as helium, hydrogen and sulfuric acid. In addition, any bending of the microwaves is an effective barometer of the atmospheric pressure, Steffes says.

"Sulfuric acid vapor, for instance, has a very unique frequency signature," Steffes says. "Using the Pioneer orbiter, we could resolve the amount of sulfuric acid vapor to an accuracy better than 10 parts per million.

"Sulfuric acid vapor is like water in the Venus atmosphere. The clouds are of sulfuric acid vapor," he says.

Like house painters trying to match the proper pastel hue, Steffes and his assistants double-check their interpretations of the radio data by mixing gas samples in a stainless steel pressure chamber and bombarding it with microwaves. They continue to adjust the mix until the microwave distortion matches that actually measured from the orbiting spacecraft.

To simulate the furnace-like surface temperatures of Venus, they cooled the laboratory pressure chamber to an oven at 600 degrees. Now, to simulate the frigid temperatures of the outer planets, Steffes is using a special freezer that can lower the chamber temperature to less than 125 degrees below zero.
July 24, 1986

Congressman Wyche Fowler, Jr.
1210 Longworth House Office Building
Washington, DC 20515

Dear Congressman Fowler:

As a strong supporter of the Space Sciences and Solar System Exploration over the past years, I thought I would take this opportunity to alert you to our concerns regarding the current appropriations debate regarding the NASA budget. As was the case with the FY 1985 budget, when I previously contacted you (March 7, 1984), attempts are currently being made to substantially cut the Research and Analysis Program within the planetary exploration budget. These attempts are probably related to the "crowding out" effect which you referred to in your March 9, 1984 letter to Congressman Michael A. Andrews, when you expressed a fear that programs like the Space Station might place such strong funding constraints on the agency that the small, potentially vulnerable, yet highly valuable, space science programs might disappear. A large portion of such funding is provided to university scientists to support research at the highest levels of excellence.

While we all are aware of the national requirements for budget restraint, the future of U.S. leadership in this field, which you have characterized as "prestigious and technology-expanding", lies in the elements of basic research and analysis, which involve students and faculty at American universities. Your support of such research in the past has been greatly appreciated, as has been your support of such issues as "peer review". We hope that it will continue both now, and in your future career, be it in the House or the Senate.

Sincerely,

Paul G. Steffes
Assistant Professor

A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA
AN EQUAL EDUCATION AND EMPLOYMENT OPPORTUNITY INSTITUTION
Appendix 5.1

GREEN BANK
OBSERVING APPLICATION
COVER SHEET

SEND TO: Director NRAO Edgemont Road, Charlottesville, Va. 22901

1 Date: April 29, 1986
2 Title of Proposal: Observations of the Venus Microwave Emission at 1.9 and 2.25 cm.

3 Authors | Institution | Who Will Observe? | Grad Student? | Observations for PhD Thesis? | Anticipated PhD Year
Paul G. Steffes | School of Electrical Engr. Georgia Institute of Tech. | Steffes & Gurski | S. (no) | G. (yes) possible | 1990

4 Contact Author
for Scheduling: Paul G. Steffes
Address: School of Electrical Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0254

5 Telephone: (404) 894-3128

6 Telescope (check one): ☑ 140" ☑ 300"

7 Scientific Category: ☑ planetary, ☑ solar, ☑ stellar, ☑ galactic, ☑ extragalactic

8 Mode: ☑ line, ☑ continuum, ☑ pulsar, ☑ VLBI, ☑ other (specify):

9 Receiver: (please consult Front End Box Status Sheet):
Cassegrain - Two Receivers (2 cm - upconverting Maser)

10 Frequency (or range of frequencies) (include test lines):
13.3 GHz and 15.8 GHz (one channel for each)

11 Sessions/Days Requested: ☑ 4-6

12 LST Range:
4-6 Local Transit ±2 hours

13 Abstract (do not write outside this space):
Observations of the planet Venus (and reference sources) at 13.3 GHz and 15.8 GHz (2.25 cm and 1.9 cm) are proposed in order to confirm the presence of a predicted "dip" in the continuum emission spectrum at 2.25 cm due to sulfuric acid vapor in the Venus atmosphere. The measurement would also provide a planet-wide average of the sulfuric acid vapor abundance in the sub-cloud region of the Venus atmosphere, a critical factor for properly modelling and understanding the Venus atmosphere.

Please attach a summary not to exceed 1000 words which contains the following information:

1) Scientific justification;
2) Observing strategy;
3) Special needs or requirements (hardware and software);
4) Source list with coordinates. In cases where such list exceeds 50 sources, a precise definition of the observational sample may be substituted.

After your proposal is scheduled, the contents of this cover sheet become public information (supporting documents are for refereeing only).
Observations of the Venus Microwave Brightness at 1.9 and 2.25 cm.

Proposed by

Paul G. Steffes, Assistant Professor
School of Electrical Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0250
(404) 894-3128

As viewed from Earth, the planet Venus is the third strongest radio source in the sky, behind the sun and the moon. As a result, it is often used as a test target for new microwave antennas. It is surprising, however, that a large amount of uncertainty still remains as to the nature of the Venus microwave emission spectrum between 1 and 22 cm. Even more surprising is the fact that no results of observations of the microwave spectrum between 2.07 and 3 cm have ever been published.

As part of a NASA-sponsored program for making laboratory measurements of the microwave properties of simulated planetary atmospheres, it has been found that the microwave absorption from gaseous sulfuric acid exhibits a peak at the 2.25 cm wavelength. This unexpected absorption behavior from a key constituent in the Venus atmosphere is quite surprising in that previously it had been believed that all microwave absorbers would exhibit absorption with a frequency-squared dependence under the high atmospheric pressures which characterize the Venus atmosphere. (For further discussion, see Steffes, 1986.) As a result, we have developed a model for the microwave emission spectrum of Venus (see attached figure), using our newly-obtained laboratory results for gaseous H₂SO₄, and previous laboratory results for SO₂ and CO₂, which provides a better fit to the ensemble of Venus microwave observations than any model yet proposed. Inspection of the model shown in Figure 1 shows a very good correlation with previously observed brightness temperatures in the 1 to 10 cm wavelength range, when a sulfuric acid vapor abundance of 15 ppm is assumed.

The most interesting aspect of our model for the Venus microwave emission spectrum lies in the 2 to 3 cm wavelength range. As can be seen from Figure 1, a noticeable dip in brightness temperature should exist between 2 and 3 cm. Unfortunately, few measurements of the Venus microwave emission have been made in this wavelength range. Two measurements have been made of the emission near 2 cm. The first, by McCullough and Boland (1964), was 500 ± 75 K at a wavelength of 2.07 cm. The second, by Pollack and Morrison (1970), was 495 ± 35 K at the 1.94 cm wavelength. Both are in good agreement with the model, assuming a gaseous H₂SO₄ abundance of 15 ppm. The most interesting wavelengths to observe would be in the 2.2 to 2.3 cm range, where a strong dip in the microwave emission, related to the abundance of gaseous H₂SO₄, should occur. Unfortunately, no observations have been published for these wavelengths. Since a high sensitivity receiver system capable of operation in this wavelength range current exists at NRAO, we are proposing such an observation.
The proposed observation would require the use of the NRAO 140-foot telescope, using the "Cassegrain, two-receiver" configuration. The two receivers used would be the 12.0-16.2 GHz upconverter/maser systems which are currently being completed. We propose to observe simultaneously at two frequencies: 13.3 GHz (2.25 cm) and 15.8 GHz (1.9 cm) in order to confirm the existence of the predicted "dip" in the continuum spectrum by observing the "differential" emission at the two frequencies, and would then attempt to make measurements of the absolute brightness temperatures by comparison with between 5 to 10 references sources in the area. In order to minimize difficulties with beam size correction and source size correction, we propose to observe Venus when it is fairly close to superior conjunction, yet still clear of the sun (either May/June 1987 or November/December 1987). Similarly, we hope to use weaker point sources as our Venus calibrators in order to minimize any beam-size correction difficulties. We propose to observe over a 4 to 6 day period with the daily observations of Venus and the references consuming approximately 4 to 6 hours.

The successful completion of the proposed observation will have several benefits:

1. Two high-accuracy data points will be added to our current knowledge of the Venus continuum microwave spectrum.

2. It will either confirm or refute the new model for the Venus microwave emission developed by Steffes (1986) (also, see Figure 1) in which sulfuric acid vapor and CO₂ are most responsible for the spectral shape at wavelengths longward of 1.8 cm.

3. Assuming the laboratory-based model by Steffes (1986) is confirmed, the resulting 2.25 cm brightness measurement should provide a planet-wide average of the sulfuric acid vapor abundance in the sub-cloud region of the Venus atmosphere, a critical factor for properly understanding and modelling the Venus atmosphere. It should likewise be noted that because of the instrumental limitations of in situ probes, this may become the major technique for determining gaseous H₂SO₄ abundance in the Venus atmosphere.

Reference:

Figure 1: Existing measurements of the microwave emission from Venus. The solid line represents the model when a 15 ppm abundance of gaseous H₂SO₄ is assumed below the main cloud layer. The dashed line represents the model when a 30 ppm abundance is assumed. Both models assume abundances for CO₂, H₂O, and SO₂ of 96 percent, 100 ppm, and 150 ppm, respectively.
EVALUATION OF THE MICROWAVE SPECTRUM OF VENUS IN THE 1.2-22 CENTIMETER WAVELENGTH RANGE BASED ON LABORATORY MEASUREMENTS OF CONSTITUENT GAS OPACITIES

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ABSTRACT

The microwave spectrum of Venus is reviewed in light of new laboratory measurements of the microwave-absorbing properties of its gaseous atmospheric constituents under simulated conditions for the Venus atmosphere. In particular, our laboratory measurements in the wavelength range 1.2-22.3 cm show that gaseous H$_2$SO$_4$ and CO$_2$ are the predominant microwave absorbers at wavelengths longer than 1.8 cm (frequencies below 17 GHz), and that SO$_2$ and CO$_2$ are the predominant absorbers at wavelengths from 1.2 to 1.8 cm. As a result, it is concluded that measurements of Venus emission/opacity at longer wavelengths (frequencies below 18 GHz) are only loosely correlated with measurements at shorter wavelengths. We also use these measurements in the development of a new model for the Venus microwave emission spectrum. The new model correlates quite well with the ensemble of microwave observations of Venus when abundances of H$_2$, SO$_2$, and gaseous H$_2$SO$_4$ are assumed to be 100 ppm, 150 ppm, and 15 ppm, respectively. We also discuss how variations in SO$_2$ abundance from 80 ppm to 200 ppm, and variations in gaseous H$_2$SO$_4$ abundance from 15 ppm to 30 ppm can explain the observed variations in 1.35 cm and 13 cm opacity, respectively. We conclude by suggesting additional observations of Venus, especially in the 2-3 cm wavelength range, in order to better evaluate the role played by gaseous sulfuric acid in the Venus atmosphere.


1. INTRODUCTION

The microwave emission spectrum of Venus in the 1.2-22 cm wavelength range has been measured and studied for nearly 30 yr. A number of works (e.g., Barrett and Staelin 1965; Muhleman, Orton, and Berge 1979; Kuz'min 1983) have summarized observations and have developed model spectra based on such observations. The microwave opacity profiles which have been inferred from these models have likewise been studied carefully in order to set limits on the abundances of microwave absorbing constituents (see, for example, Ho, Kaufman, and Thaddeus 1966 or Janssen and Klein 1981). The accuracy of such abundance estimates for microwave absorbing constituents depends critically on the availability of reliable information regarding the microwave absorbing properties of potential constituents under the conditions of the atmosphere being studied. To this end, laboratory measurements were made of the microwave absorption from potential constituents under simulated conditions for the middle atmosphere of Venus (pressures up to 6 atm): from SO$_2$ and SO$_2$ (Steffes and Eshleman 1981), and from gaseous H$_2$SO$_4$ (Steffes and Eshleman 1982 and Steffes 1985). These measurements were conducted at the 3.6 cm and 13.4 cm wavelengths in order to be directly applicable to opacity profiles obtained from radio occultation measurements. Previous to these measurements, it was assumed that the frequency dependence of the opacity from microwave-absorbing constituents, under the pressure conditions of the subcloud region of the Venus atmosphere, was simply $f^2$. (Most profiles of the radio opacity at Venus, derived from either radio occultation or radio astronomical observations, place the bulk of the opacity at altitudes below the cloud base altitude, about 48 km.) Such an assumption would preclude the need for laboratory measurements of absorption properties over a wide spectral range. While this
assumption seemed to be correct for SO₂ in a simulated Venus atmosphere (see StefTes and Eshleman 1981), it was shown not to be valid for gaseous H₂SO₄ in a simulated Venus atmosphere at wavelengths between 13.4 and 3.6 cm (Steffes 1985). As a result, we have performed further measurements of the microwave absorption spectrum of gaseous H₂SO₄ in a CO₂ atmosphere at selected frequencies between 1.34 GHz (23.3 cm wavelength) and 23.6 GHz (1.2 cm wavelength) under simulated conditions for the middle atmosphere of Venus (total pressures from 1 to 6 atmospheres). The results of these measurements significantly change the ways in which the Venus microwave emission spectrum is interpreted. We have used these results to develop a model for the microwave emission spectrum of Venus, which correlates quite well with the ensemble of microwave observations of Venus. We conclude by suggesting further observations and analysis of the Venus microwave emission spectrum, especially in the less frequently observed 2-3 cm wavelength range.

II. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of gaseous H₂SO₄ in a CO₂ atmosphere is similar to that used previously by StefTes (1985). As can be seen in Figure 1, the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of two cavity resonators at particular resonances from 1.34 GHz to 23.6 GHz. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high-resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

\[ \alpha = (Q_c^{-1} - Q_l^{-1}) \frac{1}{\lambda}, \]

where \( \alpha \) is absorptivity of the gas mixture in nepers km⁻¹. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 neper km⁻¹ = 2 optical depths per km [or km⁻¹] = 8.686 dB km⁻¹, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common [logarithmic] form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.) \( Q_c \) is the quality factor of the cavity resonator when the gas mixture is present, \( Q_l \) is the quality factor of the cavity resonator in a vacuum, and \( \lambda \) is the wavelength (in km) of the test signal in the gas mixture.

In order to obtain a gas mixture with a sufficient amount of H₂SO₄ vapor so that the microwave absorption is detectable, the system must be operated at temperatures exceeding 450 K. While this is suboptimal in that the temperatures at altitudes from 35 to 50 km (where both radio occultation and radio astronomical experiments have detected microwave opacity) range from 350 to 450 K, temperature dependencies measured for similar gases (such as SO₂) can be used to estimate temperature effects in that range. Two different approaches are used to infer H₂SO₄ vapor pressure. With the first, the volume of liquid sulfuric acid which is vaporized to generate the gaseous H₂SO₄ is determined to a high accuracy (up to ±0.005 ml). It is then possible to compute an upper limit for the partial pressure of gaseous H₂SO₄ using the ideal gas equation, the measured change in liquid volume, and published densities for H₂SO₄ liquid. The second method for accurately determining amounts of liquid-derived vapors measures the refractivity of those vapors. Since the index of
refraction (relative to unity) is proportional to the vapor abundance, the system's ability to accurately measure the refraction of such vapors can also be used to infer the relative vapor abundance and/or pressure. Note that it is not yet possible to use this approach for accurate determination of the absolute vapor pressure from gaseous H$_2$SO$_4$, since accurate refractivity data for gaseous H$_2$SO$_4$ is not currently available. While neither method is able to accurately resolve dissociation of H$_2$SO$_4$ into H$_2$O and SO$_3$, an upper limit of H$_2$SO$_4$ vapor abundance can be inferred, using the first method, which is accurate to ±2% at a temperature of 500 K.

As shown in Figure 1, a flask is filled with a precisely known volume of liquid sulfuric acid (99% by weight) at room temperature. For nearly all of the experiments, the initial volume used was 2.5 ml (measured at room temperature). Smaller quantities were also tried, with no significant difference in results. The volume measurements are made using a 1 ml syringe with 0.01 ml graduations. For volumes less than 1 ml, repeatable accuracies of better than 0.005 ml have been obtained. The entire system is then heated and allowed to thermally stabilize at the chosen experimental temperature (approximately 575 K), which requires approximately 6 hr. The temperature is monitored using a thermocouple which is placed in the center of the pressure vessel, near the microwave resonator, which is one of the coldest places in the system.

Thermal equilibrium within the system is achieved by preheating the system for 6-8 hr before beginning the experiments. Since the thermal time constant is approximately 2 hr 15 minutes, this assures a relatively constant temperature within the chamber after heating. Another technique used in monitoring the heating of the resonator within the chamber is to monitor the resonant frequencies of the resonator. As the resonator is heated, the resonant frequencies drop due to thermal expansion. Thus, when thermal stability is reached, the resonant frequencies likewise stabilize. After thermal stability is reached, a vacuum is drawn in the pressure vessel containing the microwave cavity resonator, and the bandwidth and center frequency of the resonances are then measured. For this experiment, resonances at 1.34 GHz (22.3 cm), 2.24 GHz (13.4 cm), 8.42 GHz (3.6 cm), 13.23 GHz (2.26 cm), 21.63 GHz (1.38 cm), and 23.64 GHz (1.27 cm) were used. A valve is then opened which allows the sulfuric acid vapor eluting from the flask to fill the pressure vessel (0.031 m$^3$ of open volume with resonator in place) and reach vapor pressure equilibrium with the liquid H$_2$SO$_4$. Note that all components which contact the gaseous sulfuric acid are maintained at the same temperature as the flask, so as to avoid condensation. In addition, because of the high temperatures and corrosive vapor involved, all tubing, valves, and the pressure chamber itself are fabricated from stainless steel. Gaskets and cables are fabricated from either viton or PTFE. These steps not only ensure the survival of components under the test conditions but also reduce possible reactions of the components with the sulfuric acid vapor and possible outgassing of vapors related to the materials from which the components are fabricated.

As H$_2$SO$_4$ vapor fills the chamber, changes in the resonance center frequency are observed. These changes, which reach over 400 kHz at the 13.4 cm resonance, are related to the H$_2$SO$_4$ vapor abundance. After approximately 10 minutes, the frequency shift ceases, as vapor pressure equilibrium is reached. The valve to the reservoir flask is then closed, and CO$_2$ is admitted to the chamber containing the H$_2$SO$_4$ vapor. For this experiment, a total pressure of 6 atm was used. The CO$_2$ gas is admitted to the chamber at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. The bandwidth of each response is then measured
and compared with its value when the chamber was evacuated in order to determine the absorptivity of the CO$_2$/H$_2$SO$_4$ gas mixture at 6 atm total pressure. The total pressure is then reduced by venting, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. After the system has been allowed to cool, the volume of the remaining sulfuric acid liquid (at room temperature) is measured and compared with the initial volume measured (at room temperature) in order to set an upper limit on H$_2$SO$_4$ vapor present in the gas mixture tested. This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure dependence is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency dependence of the absorptivity from a CO$_2$/H$_2$SO$_4$ mixture are likewise immune to mixing ratio uncertainty, as long as foreign-gas broadening predominates.

Since the overall equipment configuration and experimental approach for these measurements is similar to that used by Steffes (1985) for measurement of the microwave (3.6 cm and 13.4 cm) absorptivity and vapor pressure behavior of gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere, it is not surprising that the results from these experiments for the 3.6 cm and 13.4 cm opacity, and for the vapor pressure from H$_2$SO$_4$ agree quite closely with those previously obtained. However, the extended spectral coverage of these measurements (1.2-22.3 cm) presents much needed results, especially for wavelengths shortward of 3 cm. In fact, the results indicate that simple extrapolation of longer wavelength results can lead to significant errors, even with total pressure as high as 6 atm.

III. MEASUREMENT RESULTS

Measurements of the microwave absorption from gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere were conducted at a temperature of 575 K. This resulted in the evaporation of a volume of 99.0% concentration (by weight) liquid sulfuric acid which corresponded to an H$_2$SO$_4$ vapor pressure of 2.2 x 10^{-2} atmospheres in our pressure vessel. (We assumed that 47% of the vaporized H$_2$SO$_4$ dissociated to form SO$_3$ and H$_2$O, as computed by Gmitro and Vermeulen 1964 for 99% solutions.) This value for H$_2$SO$_4$ vapor pressure agreed quite closely with the value obtained in earlier measurements (see Steffes 1985). Since the same earlier measurements indicated that microwave opacity from gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere increased with frequency, we made our initial high-frequency measurements of absorptivity at wavelengths around 1.3 cm, expecting large opacities. Much to our surprise, we were unable to detect opacity in this wavelength range from the H$_2$SO$_4$/CO$_2$ mixture. The experiment was repeated several times, with equipment being rechecked, but the results were still the same. Since the refractivity of the gaseous H$_2$SO$_4$ and CO$_2$ were easily measured at all wavelengths as they entered the pressure chamber, it was clear that the gases were indeed present in the quantities expected. In fact, the absorptivity measured at the 13.4 cm and 3.6 cm wavelengths agreed quite well with previous measurements by Steffes (1985). It was
therefore clear that even with total pressures as high as 6 atm, that mixtures of gaseous H$_2$SO$_4$ and CO$_2$ had a unique microwave absorption spectrum which differed significantly from a simple $f^2$ dependence. It should also be noted that the maximum value of the collision-induced absorption from pure CO$_2$ as determined by Ho, Kaufman, and Thaddeus (1966) could not exceed 0.03 dB km$^{-1}$ under the conditions of these experiments. Since this is far below the threshold sensitivity of our system, it had no affect on the measurements.

Our measurements results for the microwave absorption from gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere at 575 K are shown in Figure 2. The data points presented are for an H$_2$CO mixing ratio of 0.4% and were obtained for total pressures of 6 atm and 1 atm. Best-fit curves for the absorption spectra from 1.3 GHz (22.5 cm) to 25 GHz (1.2 cm) at pressures of 6 atm and 1 atm are also shown. Inspection of these results reveals three major results:

1. The first major result is the extremely low absorptivity observed in the 1.2-1.8 cm wavelength range. For example, measurements made at 21.63 GHz (1.38 cm wavelength) show an opacity for 0.4% mixture of gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere (with a total pressure of 6 atm and temperature of 575 K) of less than 9 dB km$^{-1}$. This is far below the opacity predicted by using previous measurements at 2.2 GHz and 8.4 GHz and assuming a simple $f^2$ dependence such as exhibited by SO$_2$ and CO$_2$. In fact, this even implies that using the measured frequency dependencies for the microwave absorption from H$_2$SO$_4$ in a CO$_2$ atmosphere in the 2.2-6.4 GHz range to predict absorption at 21.6 GHz (1.38 cm wavelength) would result in overstating the absorption at 1.38 cm by at least a factor of 2.

Such behavior implies that the contribution of gaseous H$_2$CO to the overall non-CO$_2$ opacity inferred from radio astronomical observations at the 1.35 cm wavelength is minimal. Such a result is not surprising in light of the calculations done by Janssen and Klein (1981) which attribute nearly all of the non-CO$_2$ opacity at 1.35 cm in the Venus atmosphere to SO$_2$. This behavior is likewise consistent with the calculated H$_2$SO$_4$ resonance frequencies computed by Poynter (R. Poynter, personal communication. Note that the results of the resonance calculations are shown in Cimino 1982.). These calculations are based on rotational constants computed using measurements of H$_2$SO$_4$ resonances in the 60-120 GHz range by Kuczkowski, Suenram, and Lovas (1981). It is significant that there is a notable absence of H$_2$SO$_4$ resonances in the 15-30 GHz frequency range, which would be consistent with our measurements.

2. The second major result is the discovery of a peak in the absorptivity from gaseous H$_2$SO$_4$ at a wavelength of approximately 2.2 cm, even at pressures as high as 6 atm. This does a lot to explain why observations of the Venus brightness temperature at 2 cm (495 K—from Pollack and Morrison 1970), which were initially felt to be inconsistent with measurements at 1.35 cm (474 K—from Muhleman, Orton, and Berge 1979), can indeed be consistent, since there is at least one absorber whose opacity decreases with increasing frequency, in that wavelength range. In fact, we have developed a model for the microwave emission spectrum of Venus, using our laboratory results for gaseous H$_2$SO$_4$, and previous results for SO$_2$ and CO$_2$ which provides a better fit to the ensemble of Venus microwave observations than any model yet proposed (see § IV). A computation of the microwave absorption spectrum from H$_2$SO$_4$ made by Allen, which is presented in Cimino (1982), shows significantly less absorption from H$_2$SO$_4$ in this same frequency range than we have actually measured. This can be explained by the fact that a pressure-hardened
linewidth parameter of 7.2 MHz was assumed for gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere. It appears that using a smaller broadening parameter would result in a spectrum which would be more consistent with our measurements.

3. The third major result is the low absorptivity measured at 22.3 cm. This is important since the well-known problem of the fall-off of brightness temperatures at wavelengths longward of 20 cm (see, for example, Muhleman, Orton, and Berge 1979) is still unsolved, and might have been explained by a large opacity from a gaseous constituent. Our negative result indicates that no atmospheric constituent can be responsible for this effect.

In Figure 3, we compare the microwave absorptivity from 15 ppm gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere with that from 150 ppm SO$_2$ in a CO$_2$ atmosphere, under pressure and temperature conditions corresponding to an altitude of 35 km in the Venus atmosphere. The opacity from SO$_2$ is computed from the results of Steffes and Estleman (1981). The opacity from gaseous H$_2$SO$_4$ is as presented in Figure 2 for a pressure of 6 atm, however, an estimated temperature dependence of $T^{-2}$ was used in order to determine the effects of the colder temperature at the 35 km altitude of the Venus atmosphere (450 K vs. 575 K used in the laboratory). Likewise, since the microwave absorption from a minor constituent (number mixing ratio less than 10%) is generally directly proportional to its abundance (either by number or by volume), we assume a linear dependence with mixing ratio. The SO$_2$ mixing ratio (150 ppm) was picked based on situ measurements (see, for example, Oyama et al. 1980 or Gel'man et al. 1979). The H$_2$SO$_4$ mixing ratio (15 ppm) corresponds to the minimum abundance necessary so as to reach saturation under Venus atmospheric conditions at an altitude at or above 48 km, the highest measured cloud base altitude (see Knollenberg et al. 1980). Note we assume H$_2$SO$_4$ saturation vapor pressure characteristics as described by Steffes (1984). Inspection of Figure 3 shows that while radio occultation and radio astronomical measurements at wavelengths longer than 1.8 cm detect absorption at the 35 km altitude which is predominated by gaseous H$_2$SO$_4$, opacity at this altitude measured by radio astronomical observations in the 1.2-1.8 cm wavelength range is predominated by SO$_2$.

At other altitudes, the frequency above which opacity from SO$_2$ (150 ppm) predominates over that from gaseous H$_2$SO$_4$ (15 ppm) is essentially the same. It should be noted that this applies only to altitudes from 30 km to 50 km, since most chemical models of the Venus atmosphere require the dissociation of gaseous H$_2$SO$_4$ at altitudes below 30 km (see, for example, von Zahn et al. 1983), and since the sulfuric acid vapor condenses out at altitudes above 48 km. It should also be noted that at altitudes below 30 km in the Venus atmosphere, the collision-induced microwave absorption from CO$_2$ (Ho, Kaufman, and Thaddeus 1966) exceeds that from expected abundances of SO$_2$ (150 ppm) or H$_2$SO$_4$ vapor (below 10 ppm), regardless of wavelength.

Thus, our measurements suggest that at wavelengths longer than 1.8 cm, the opacity from the expected quantities of gaseous H$_2$SO$_4$ (15-30 ppm) would predominate in the 30-50 km altitude range in the Venus atmosphere, while the opacity from expected quantities of SO$_2$ predominates at wavelengths from 1.2 to 1.8 cm. At wavelengths shortward of 1 cm, the relative predominance of the opacity from either gaseous H$_2$SO$_4$ or SO$_2$ is not as clear, since both have large numbers of rotational resonances throughout the millimeter-wavelength range. Laboratory measurements of the millimeter-wave absorption properties of constituent gases under simulated conditions for the Venus atmosphere could be a useful tool for
interpreting the millimeter-wave spectrum of Venus.

IV. MODELS FOR THE MICROWAVE EMISSION SPECTRUM OF VENUS

Several models for the microwave emission spectrum of Venus have been developed, based on numerous observations, in attempts to infer constituent abundances and atmospheric structure. However, the lack of reliable data on both the identities and microwave absorbing properties of many of the atmospheric constituents cast an aura of uncertainty over the resulting inferred atmospheric models. When Pollack and Morrison (1970) attempted to explain the microwave emission spectrum of Venus, they attributed all microwave atmospheric opacity to CO₂, N₂ gas, gaseous H₂O, and dust. It was found that in order to match the observed brightness temperature at the 1.94 cm wavelength, an H₂O vapor abundance ranging between 3500 ppm and 10,000 ppm was required. This value far exceeds any in situ measurement of water vapor abundance made by any of the subsequent atmospheric probes. In fact, current models of the Venus atmosphere place water vapor abundances below the cloud layers in the 100 ppm range (see Zahn et al. 1983). In addition, the accompanying opacity at 1.35 cm from such quantities of water vapor would result in a 1.35 cm brightness temperature which would be notably below those measured (see Janssen and Klein 1981).

Muhleman, Orton, and Berge (1979) used an alternative approach for modeling the opacity from atmospheric constituents. Like Pollack and Morrison (1970), they found that opacity from CO₂ alone could not explain radio observations of the planet; that is, additional sources of radio absorption were required. However, instead of attributing this absorption to a specific constituent with known absorption properties, it was assumed that all sources of additional opacity would exhibit microwave opacity whose pressure, temperature, and frequency dependencies would be the same as those for CO₂. It should be emphasized that this was a perfectly reasonable approach for modeling microwave opacity in the Venus atmosphere previous to the Pioneer-Venus mission, since a large number of potential absorbing constituents might have existed, the microwave absorption properties for which, under Venus atmospheric conditions, were unknown. Similarly, since the constituents whose opacity had been measured under simulated laboratory conditions for the Venus atmosphere (CO₂, H₂O, and N₂—see Ho, Kaufman, and Thaddeus 1966) had all exhibited the same basic pressure, temperature, and frequency dependence, this assumption seemed to be reasonable; yet the authors recognized and stated the possibility for significant error.

As with previous models for atmospheric opacity in the Venus atmosphere, the model used by Muhleman, Orton, and Berge (1979) resulted in a theoretical microwave emission spectrum which agreed quite well with the actual observations at certain wavelengths, but poorly at others. While some of these discrepancies may have been due to calibration errors in the observed data, an opacity frequency dependence other than 1 would be required in order to better match the observed brightness temperatures at other wavelengths, especially near 2 cm. It was also pointed out that such a difference in frequency dependence was also required in order for the opacity models in the 1.35 cm wavelength range to be able to match observed opacity at 3.6 cm and 13 cm as measured by radio occultation experiments. This was likewise recognized by Janssen and Klein (1981).

As shown in Figure 4, we have developed a model for the microwave emission spectrum from Venus based on the laboratory measurements of opacities of the potential constituents. Computations of the modeled brightness temperatures were
made by OQ£/Gurski (private communication) and assume that the primary absorbing constituents of the Venus atmosphere are $\text{CO}_2$, $\text{SO}_2$, and gaseous $\text{H}_2\text{SO}_4$. While some microwave opacity can also be attributed to gaseous $\text{H}_2\text{O}$ and the cloud layers themselves, the contribution from the expected abundance of gaseous $\text{H}_2\text{O}$ (in the 100 ppm range below the clouds—see von Zahn et al. 1983), and from the cloud layers as measured by Knollenberg and Hunten (1979) and by Ragent and Blamont (1979), would be very small. For our model, we assume an atmosphere with a 96% abundance of $\text{CO}_2$, and a temperature/pressure structure as measured by the Pioneer-Venus probes (Seiff et al. 1980). The opacity from $\text{CO}_2$ is computed using the laboratory-based expression for $\text{CO}_2$ microwave opacity from Ho, Kaufman, and Thaddeus (1966) and assuming a $\text{CO}_2$ abundance of 96%. We assume a $\text{SO}_2$ abundance which is essentially zero above the main cloud layer (48-50 km) and 150 ppm below the main cloud layer down to the surface. The opacity from $\text{SO}_2$ is computed from the results of Steffes and Eshleman (1981). For gaseous $\text{H}_2\text{SO}_4$, we use the results from § III and assume a uniform abundance between the 30 km altitude and the cloud base (approximately 48 km). Below 30 km, we assume dissociation of gaseous $\text{H}_2\text{SO}_4$ into $\text{SO}_2$ and $\text{H}_2\text{O}$ (which are both relatively transparent in the abundances expected) and above 48 km, we assume condensation begins. In Figure 4, we present microwave emission spectrum for two different abundances of gaseous $\text{H}_2\text{SO}_4$: 30 ppm and 15 ppm. These abundances are near the upper and lower limits for the gaseous $\text{H}_2\text{SO}_4$ abundance required to account for the 13 cm opacity measured by radio occultation experiments (see Steffes 1985). Also plotted are actual observed brightness temperatures, as given by Pollack and Morrison (1970), Kuz'min (1983), and Muhleman, Onion, and Bering (1979). It should be noted that a constant surface microwave brightness of 640 K is assumed, hence the apparent variation between the model and observed emission at longer wavelengths.

Inspection of our model shows a very good correlation with observed brightness temperatures in the 1-10 cm wavelength range, when a sulfuric acid vapor abundance of 15 ppm is assumed. However, certain aspects of the spectra bear special consideration. The first is the 1.2-1.5 cm wavelength range. A great number of brightness measurements have been made in this wavelength range, especially at the 1.35 cm wavelength (the center of the $\text{H}_2\text{O}$ absorption band). These measurements range from 550 ± 26 K (see Janssen 1972), down to 456 ± 39 K (Law and Staelin 1968). While some of the resulting variations in the 1.35 cm atmospheric opacity required to explain these brightness temperature variations are probably due to interpretive or calibration inaccuracies, since the observations were made with relatively primitive receivers and since there is substantial uncertainty in the 1.35 cm flux calibration scale, they may suggest significant temporal changes in atmospheric opacity, such as might be induced by volcanism (see, for example, Esposito 1985, or Prinn 1984). A question then arises as to which constituents could be responsible for such opacity variations. Inspection of Figure 4 shows that assuming abundances of 150 ppm $\text{SO}_2$ and 15 ppm $\text{H}_2\text{SO}_4$, our model predicts a 1.35 cm brightness temperature of 490 K. Even if the $\text{H}_2\text{SO}_4$ abundance were doubled to 30 ppm, the resulting reduction in the 1.35 cm brightness temperature would be only 25 K. This is not surprising in light of the relatively low opacity exhibited by $\text{H}_2\text{SO}_4$ at these wavelengths. Thus, we conclude that the variations in 1.35 cm brightness cannot be attributed to variations in the abundance of gaseous $\text{H}_2\text{SO}_4$, since the required abundance variations would far exceed allowable limits for $\text{H}_2\text{SO}_4$ abundance set by sulfuric acid vapor satura-
tion or 13 cm opacity measurements from radio occultation experiments (see Steffes 1985).

An alternative explanation for opacity variations would be variations in the abundance of SO$_2$. If the SO$_2$ abundance of our model were increased from 150 to 200 ppm (a 33% increase), the 1.35 cm brightness (assuming a constant H$_2$SO$_4$ abundance of 15 ppm) would go from 490 K down to 460 K, well within the error bars of the Law and Steinel (1968) measurement, while not seriously affecting the 13 cm opacity or the emission brightness at wavelengths longer than 1.8 cm. Similarly, reducing the SO$_2$ abundance to 80 ppm results in a brightness temperature of approximately 525 K which is within the error bars of the Janssen (1972) measurement. It should be emphasized that these apparent long-term variations in opacity may be partially due to calibration inaccuracies; however, the required long-term variations in SO$_2$ abundance required to explain these opacity variations would be of the same order as the long-term variations in upper atmospheric SO$_2$ abundances as observed by Esposito (1985).

The second interesting aspect of our model for the microwave emission spectrum lies in the 2-3 cm wavelength range. As can be seen from Figure 4, a noticeable dip in brightness temperature should exist between 2 and 3 cm. Unfortunately, few measurements of the Venus microwave emission have been made in this wavelength range. Two measurements have been made of the emission near 2 cm. The first, by McCullough and Boland (1964), was 590 ± 75 K at a wavelength of 2.07 cm. The second, by Pollack and Morrison (1970), was 495 ± 35 K at the 1.94 cm wavelength. Both are in good agreement with the model, assuming a gaseous H$_2$SO$_4$ abundance of 15 ppm. The most interesting wavelengths to observe would be in the 2.2-2.3 cm range, where a strong dip in the microwave emission, related to the abundance of gaseous H$_2$SO$_4$, should occur. Unfortunately, no observations have been published for these wavelengths. Since high-resolution, high-sensitivity receiver systems capable of operation in both this wavelength range and at 1.35 cm do exist, we suggest that long-term observation of Venus microwave emission could be made so as to characterize variations of the SO$_2$ abundance (1.35 cm) and gaseous H$_2$SO$_4$ abundance (2.2 cm), both on a planet-wide basis, and on a localized basis, as limited by instrument spatial resolution.

As mentioned, our model assumes a constant surface brightness temperature of 640 K. This simplification could be substantially improved. Muhleman, Orton, and Berge (1979) used radar reflectivity measurements in developing a better model for surface brightness but still could not match observations of emission at wavelengths longward of 20 cm. It is likely that laboratory measurements of dielectric properties of simulated surfaces, under simulated Venus atmospheric conditions, may be able to provide an understanding of this phenomenon, and should therefore be pursued. It should also be noted that the expression used for opacity from SO$_2$ was based on laboratory measurements in the 1-6 atm pressure range. This expression may underestimate the opacity from SO$_2$ in the 50-100 atm (6-10 km) range, which would result in our model understating the Venus brightness temperature, especially in the 4-6 cm wavelength range.

V. CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK

Laboratory measurements have been made of the microwave opacity from gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere, under simulated conditions for the middle atmosphere of Venus, in the 1.2-22.3 cm wavelength range. Our laboratory results have shown that the predominant microwave absorbers in the Venus atmosphere at wavelengths longer than about 1.8 cm are gaseous H$_2$SO$_4$ and CO$_2$, whereas SO$_2$ and CO$_2$ pre-
dominate at shorter wavelengths. Thus, in using these measurements to model the Venus microwave emission spectrum, we have found that gaseous H$_2$SO$_4$ and CO$_2$ are the constituents most responsible for the Venus microwave emission spectrum at wavelengths longer than about 1.8 cm, and that SO$_2$ and CO$_2$ most affect the emission spectrum between 1.2 and 1.8 cm. Our model of the Venus microwave emission spectrum correlates well with the ensemble of microwave observations of Venus when abundances of H$_2$O, SO$_2$, and gaseous H$_2$SO$_4$ below the main cloud layer, are assumed to be 100 ppm, 150 ppm, and 15 ppm, respectively. Analysis of our model shows that the significant variations observed in the 1.35 cm emission cannot be explained by allowable variations in the abundance of gaseous H$_2$SO$_4$ abundance (15-30 ppm, as limited by 13 cm opacity measurements), but could be explained by variations in the subcloud SO$_2$ abundance from approximately 80 ppm to 200 ppm.

Additional microwave observations of Venus should be conducted both at 1.35 cm and near the 2.2 cm wavelength in order to better characterize both spatial and temporal variations in abundances of atmospheric SO$_2$ and gaseous H$_2$SO$_4$. We also intend to conduct further analysis of existing observational data, in light of the laboratory results. A final critical issue to the understanding of the Venus microwave spectrum involves the surface microwave emission. Actual laboratory measurement of the microwave dielectric properties of simulated surfaces under Venus atmospheric conditions would be an important key to a better understanding of the microwave emission of the surface as well to characterizing variations in surface structure from existing and future radar measurements.

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Fig. 1.—Block diagram of the atmospheric simulator as configured for measurements of the microwave absorption of gaseous H$_2$SO$_4$ under Venus atmospheric conditions over the 1.2-22.3 cm wavelength range.

Fig. 2.—Laboratory measurements of absorptivity (dB km$^{-1}$) of gaseous H$_2$SO$_4$ in a CO$_2$ atmosphere ($T = 575$ K; H$_2$SO$_4$ mixing ratio = 0.4%). Solid curves are best-fit expressions for mixtures with total pressures of 6 atm and 1 atm.

Fig. 3.—Comparative microwave absorption from gaseous H$_2$SO$_4$ (15 ppm, solid line) and SO$_2$ (150 ppm, dashed line) at the 35 km altitude in the Venus atmosphere ($P = 6$ atm; $T = 450$ K).

Fig. 4.—Measurements of the microwave emission from Venus. The solid line represents the model when a 15 ppm abundance of gaseous H$_2$SO$_4$ is assumed below the main cloud layer. The dashed line represents the model when a 30 ppm H$_2$SO$_4$ abundance is assumed. Both models assume abundances for CO$_2$, H$_2$O, and SO$_2$ of 96%, 100 ppm, and 150 ppm, respectively.

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Figure 1
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