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13. ABSTRACT (Maximum 200 words) This project was designed to identify applications of THz technology of potential interest to the DoD and to provide a technical introduction for DARPA personnel to the underlying scientific and technical issues. The final report begins with a discussion of the physics which governs the interactions between radiation and matter in the THz, with emphasis on the differences between low pressure gases, atmospheric pressure gases, and solids. Atmospheric propagation is considered as an example. Two specific applications were developed in more detail. The first is a concept for a point detector of gases, which is based on the absolute specificity of rotational spectra in the THz. The second is a remote detection scheme which makes use of THz/IR double resonance. The former is based on well understood science and the risks for the development of such a program lie in the trade-offs against alternative solutions as a function of scenario. Although the details of application scenarios are not known to the authors, it is highly likely that some of them are very favorable. The latter is a high risk/high payoff scheme whose eventual utility depends not only upon application scenario, but also on currently unknown scientific parameters.			
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Feasibility of THz Remote Sensing for DoD Applications

A Final Report

Submitted to Lt. Col. Gernot Pomrenke

Air Force Office of Scientific Research Foundation

By

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October, 2000

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Additional input has been submitted by Dr. Simon Verghese of Lincoln Laboratory

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INTRODUCTION

A convenient organizational paradigm is that the evaluation of any spectral region for possible application can be divided into the consideration of the production and detection of radiation, the propagation/transmission of the radiation from source to detector, and the interaction of that radiation with matter. From this perspective two important attributes mark the THz and determine both the current state-of-the-art and what is ultimately possible: the difficulty of producing THz radiation and the nature of its atmospheric propagation. The former is discussed in a companion report from Simon Verghese of Lincoln Laboratory,

Also included in this report is a specific discussion of gas detection in the THz. Because the basic resolution limit for molecules is Doppler broadening (which is proportional to frequency), orders of magnitude higher resolution is possible in the THz region than in the infrared or optical. A result of this is that the complex and unique rotational 'finger-prints' of the large majority of gas phase molecules can be resolved and the observation of the THz Doppler limited spectra results in an *absolutely specific* identification in virtually all circumstances. However, to be Doppler limited in the THz, the pressure must be ~ 0.1 Torr. As a result, at atmospheric pressure THz spectral lines (as well as infrared and visible) are broadened to ~ 5 GHz and both specificity and sensitivity are lost. Any attempt to use THz techniques to detect trace gases at atmospheric pressures must address these issues.

After briefly discussing the basic physics of interactions in this spectral region and atmospheric propagation, specific examples of THz molecular spectra will be presented to illustrate specific cases of interest. Finally, a small, fast, and potentially inexpensive THz point detector that has a well-defined research/development path will be described along with a speculative, high-risk scheme for the detection of trace gases at atmospheric pressure. Because the former will operate at low pressure, its identifications of gases will be quantitative and virtually absolutely specific. Because of its use of a time resolved, double resonance scheme, the atmospheric pressure remote sensing proposal, while not absolutely specific, should have much higher specificity than would result from the usual spectroscopic gas detection schemes in the atmosphere.

I. The basic physics

We will begin with a brief review of the basic physics of the interaction of THz radiation with matter, with specific examples that impact sensors and remote sensing. Because the nature of the interactions in the condensed phase and the gas phase are fundamentally different, we will begin by discussing them separately. To a very good approximation the interactions of THz radiation with gases depend upon the properties of the rotational quantized states of molecules, while those in solids are describable classically as lossy dielectrics.

Gases:

1. Individual **atoms** in general are transparent to THz radiation. Their electronic energy level differences fall in the optical and ultraviolet and their nuclear fine and hyperfine interactions are very small and typically occur in the radio. Furthermore, there are few free gas

phase atoms in nature and this, coupled with the very small absorptions at radio frequencies, accounts for their unimportance.

2. **Molecules** have a much richer set of energy levels and interactions: electronic levels which give rise to optical and ultraviolet spectra, vibrational level (the heavy nuclei vibrating in the intermolecular potential provided by the electrons and other nuclei) in the infrared, rotational levels in the microwave, and nuclear hyperfine interactions in the radio. Because of this energy hierarchy, virtually all of the interactions in the THz region are related to rotational interactions and we will focus on these interactions.

2a. **Transition Frequencies:** Because angular momentum is quantized, rotational energy levels are discrete. **Figure 1** shows an absorption spectrum of H_2O , H_2S , CH_3Cl , CO , HCN , and HCl between 0 and 1 THz. However, because this interval has $\sim 10^6$ Doppler limited resolution elements, it is possible to plot fewer than 1/1000 of the resolution elements. As a result, this spectrum which looks crowded is, in fact, virtually empty. This will be illustrated in more detail below. Several important features can be illustrated in this figure which are general. First, the absorptions are very strong. The plot assumes a dilution to 1 ppm. If there were no atmospheric effects, the noise level of a system with 1 second of integration time would be off of the bottom of the chart. Starting at low frequency the absorption strength grows as frequency squared (For complex molecules, it is really more complicated than this, but this is more of a detail than an issue of importance for the purposes of this discussion), peaks somewhere in the THz region, and then falls exponentially. A more detailed analysis shows that the frequency of this peak is a function of the size/mass of the molecules, with the peak of a small molecule like water falling at many THz, whereas heavier gases like Sarin have peaks around 0.3 THz. This is discussed in more detail in the attached reprint from an article in *Analytical Chemistry*.

2b. **Transition Linewidths:** Of special significance to both remote sensing through the atmosphere and the use of THz radiation for molecular detection are the mechanisms which broaden the spectral lines. The two of most significance for this study are pressure broadening and Doppler broadening. Doppler broadening arises from the random kinetic motion of the molecules. For gas phase molecules near ambient temperature the ratio of the Doppler broadening to the transition frequency is $\sim 10^{-6}$ (~ 100 kHz at 100 GHz). Molecules pressured broadened in the atmosphere typically have widths of 5 GHz. Thus, molecular lines in the atmosphere have lines that are typically 5000 times broader than lines in a spectroscopic cell (typically run under Doppler broadened conditions).

2c. **Quantitative Analysis and measured linestrengths:** Because the absorption strengths of rotational transitions can be linked both experimentally and theoretically to concentration, THz rotational spectra also provide accurate quantitative measurements of gas concentrations.

2d. **Spectral complexity/specificity:** It is significant that many ($10^2 - 10^5$) rotational levels are thermally populated, whereas for vibrational and electronic levels typically only the ground state is populated near 300 K. This results in great rotational spectral complexity/specificity in the THz. **Figure 2** shows (again on a compressed scale - fewer than 1/100 of the resolution elements can be plotted) the spectrum of SO_2 between 200 and 300 GHz. Because this is a heavier species it has a much more complex and dense spectrum than any of the species shown in **Figure 1**. However, when its spectrum is blown up in **Figure 3** to show a 1 GHz region (here the Doppler width is perhaps 1/5 of the width of the plotted line) the spectrum once again appears virtually empty. **Figure 4** shows a 100 GHz region of **Figure 1**. Here, because the molecules are lighter much empty space occurs even on this compressed plot. Issues and

examples involving heavier species with denser spectra are considered in more detail in the attached article from *Analytical Chemistry*.

Thus, because in the THz so many resolution elements are available and filled by the rotational spectral of molecules, even in complex mixtures absolutely specific identification and quantitative analysis are possible at low pressure. This is a unique feature of the THz spectral region.

2e. Atmospheric Pressure: **Figure 5** shows the same gases as **Figure 1**, but broadened to the ~ 5 GHz typical of pressure broadened lines in the atmosphere. Comparison to **Figure 1** shows that the absorption strengths are essentially the same, but that the $\sim 10^6$ resolution elements have been reduced to ~ 100 . **Figure 6** is the same as **Figure 5** except that a real standard atmosphere has been included. In this graph water has been increased from 1 ppm to $\sim 1\%$ (7 Torr partial pressure) and an empirical continuum (see below) included. Note that the absorptions of the trace gases have not been reduced, but rather they are overwhelmed by the absorption of the abundant water (1%) and oxygen (20%). The impact of the real atmosphere on trace gas detection thus has two elements:

- i. The absorption that reduces the probe signal, especially at the higher THz frequencies, and
 - ii. The fluctuations in the probe signal due to atmospheric fluctuations (which are largely unknown and unstudied, but which probably occur on a meteorological time scale ($\gg 1$ ms)).
- On this time scale, these fluctuations will dominate by many orders of magnitude the system noise, which is off of the bottom of this chart by perhaps 5 decades. Below, we will propose a high risk 'double resonance' modulation scheme to overcome this problem as well as to significantly increase specificity.

3. THz compared to IR: Although not specifically an issue here, it is useful to make a brief comparison with infrared interactions:

- a. The pressure broadening is wavelength independent - thus lines which are pressure broadened have widths which are independent of spectral region.
- b. Doppler broadened lines have widths proportional to the frequency. Thus for Doppler broadened lines, THz lines are typically 100 times narrower than infrared lines.
- c. At atmospheric pressure, pressure broadening is dominant in both the THz and the IR, resulting in similar linewidths.
- d. In a point sampling system, it is advantageous to adjust the pressure so that the Doppler limit is reached. This results in about 100 times narrower linewidths for THz systems. This factor of 100 is significant because for many gases of 'detection interest' the highly specific rotational "fingerprint" which is unresolvable in the IR, becomes resolvable in the THz.
- e. Although it varies enormously by molecular species, absorption strengths are similar in the THz and the IR. The IR photons and population differences are larger, whereas the THz quantum mechanical transition moments are larger.

Solids:

We will comment only briefly on solids because from the point of view of the basic physics that impacts remote sensing in the THz they are 'uninteresting'. This is not to say that interactions with solid are 'unimportant' for remote sensing in the THz, but rather that remote

sensing depends on the complexity of the physical situation (e. g. the shape, distribution, and water content of leaves or of particles in 'dry-wall') rather than the details of quantum physics.

To a good approximation, in the THz solids are characterized by their classical Electricity and Magnetism properties (i. e. their complex dielectric constants). Metals are good reflectors, and many plastics and other non-conductors are relatively low loss dielectrics. A surprising, but illustrative example is the fact that while quartz is an extremely low loss dielectric at THz frequencies, ordinary window glass is rather lossy (~50% for 1/8"). It is also noteworthy that while in the infrared painted metals have reflective properties largely determined by the nature of the paint, in the THz most paint is transparent enough that the reflective properties of painted metals are largely that of the underlying metal.

The scattering properties of solids in the THz are important. Briefly stated, for wavelengths long in comparison to particle size, radiation propagates largely unscattered; whereas for wavelengths small in comparison to particle size, significant scattering occurs. For the classic case of atmospheric propagation, THz waves are largely unscattered by fog, dust, and smoke (which severely scatter infrared and optical radiation), but heavy rain impacts propagation well into the microwave region. It is this ability to penetrate obscurants which have led over the years to many of the applications which have been envisioned for millimeter, submillimeter, near millimeter, and terahertz technologies.

It is also probably worth noting that it is often easy to mix (both experimentally and conceptually) the effects of scattering and the effects of attenuation. As an example, consider (1) an object, (2) a scattering medium such as frosted glass, and (3) an observer. If the frosted glass is placed immediately over the object and far from the observer, little image resolution or intensity is lost. However, if the frosted glass is placed immediately in front of the observer, the image is completely lost. This has impact on schemes involving 'seeing' through clothes (easy, because the scatterer is next to the object) or walls (highly scenario dependent, because of the variable locations of the scatter between the object and the observer).

II. An Example, Atmospheric Propagation:

It is convenient that the atmospheric propagation shown in **Figure 6** is both important to this study and can serve as an excellent illustration of much of the basic physics. **Figure 7** shows a more general atmospheric propagation plot which includes infrared and optical frequencies as well as the effects of scatterers such as rain and fog. This plot was originally drawn by hand by Bobby D. Guenther of the Army's Missile Command in Huntsville in the late 1970s. Comparison of the two shows a few differences. The logarithmic plot of frequency in **Figure 7**, made it difficult to accurately hand draw the linewidths (which should stay at a constant width of ~5 GHz from the microwave through the THz). This makes the widths of the THz windows appear much narrower than they really are. Secondly, an order of magnitude was lost in the drawing process for the strongest absorbers in the THz region.

It is important to realize that the attenuation shown on the vertical of both graphs is in effect plotted as the log of a logarithmic measure. *As a result, seemingly small changes are in fact very large.*

1. The states of water: Most of the attenuation in the THz region can be attributed to water or oxygen. A large, controversial, and in many cases unreliable literature exists about the role of water and the possible forms in which it *may* exist in the atmosphere. Gas phase water consists of individual molecules which collide with other atmospheric constituents at a rate of $\sim 10^9$ /sec.

The absorption due to this water is calculated quantum mechanically as discussed above. Rain or fog consists of droplets of water. In these droplets, there is no molecular rotation. In this case, the physical size of the droplet gives rise to a scattering process which is governed by classical electricity and magnetism calculations. The basic result of this is the well known result that radiation of wavelength longer than the particle size passes through the droplet with little scattering, whereas radiation whose wavelength is comparable or shorter than the droplet size is severely scattered. The transmission in the important window regions is much less than would be calculated from well founded theory. As a result a host of mutually exclusive explanations have been put forth to explain the 'anomalous' absorption and included in this host are explanations based on other possible states of water. Most prominent among these explanations are those involving dimers, trimers, etc. Although beyond the scope of this paper, we would urge that any work undertaken related to this path of investigation be done with caution and a full review of earlier work.

2. Gas Phase Absorption: In the THz region all of the peaks of absorption shown can be attributed to either water vapor or oxygen. For a strong rotational interaction with radiation, a molecule must have a permanent electric dipole moment. Neither of the major constituents of the atmosphere N_2 or O_2 have electric dipole moments because of their symmetry. However, O_2 is an important oddity in that it is one of the few stable molecules with a *magnetic* dipole moment, which gives rise to a weaker rotational interaction.

As noted above, the strengths of molecular interactions increase roughly as the square of the frequency. It can be seen that even in the complexities of the real atmosphere that this trend is followed. Again, as noted above, there is a maximum (which for the small/light H_2O is at a fairly high THz frequency) before the exponential decline. Although complex in detail, for the purposes of this study it should be noted that all of the linewidths are due to pressure broadening and are about 5 GHz. The logarithmic plot distorts this somewhat. The linewidths of the rotational-vibrational lines in the infrared are also of similar width, but the logarithmic plot merges all of the individual lines together and each of the plotted peaks are *much* wider vibrational band contours, each of which contains many rotational lines.

3. Scattering: Also plotted in this figure are attenuations due to various rainfall rates and fog. While in principle these are straightforward classical calculations, there is enormous possible variation in the particle size distributions for fog and rain and these graphs should be considered only as indicative of the range of possible results.

III. The Detection, Identification, and Quantification of Gases via THz Techniques

The study of molecular gases was the first application of submillimeter waves and is still by far the largest application of this spectral region, having been expanded from the laboratory to include remote sensing of molecules in the upper atmosphere and the interstellar medium. The reasons for the importance of submillimeter/THz molecular spectroscopy include the very strong interactions between submillimeter radiation and molecules and the small Doppler broadening. The latter gives access to the highly specific and scientifically informative rotational energy level spectrum of gas phase molecules.

In turn, these spectroscopic interests have driven the development of most of the technology which is currently used in the submillimeter/THz region to make scientific measurements or technological observations. This is no accident because the need for sensitivity, high resolution, and above all a practical technology is very high in molecular spectroscopy.

However, it is well known that the submillimeter/THz is a difficult region technologically (primarily for the development of sources) and this difficulty has precluded its wide spread use beyond the bounds of a relatively small community driven by particular scientific or technological interests. While it is beyond the bounds of this report, it must be mentioned that for many applications the current technological limits are not placed by fundamental physical limits but rather from the small market precluding the mass production of technologies which have been known for many decades. The most notable example is the development of exquisite Backward Wave Oscillators (BWOs) by the Russians, for whom market was not a consideration.

Below we will discuss two approaches to the use of the submillimeter/THz for gas detection. The first is a **point detector**. It is firmly based on many years of submillimeter spectroscopic knowledge and provides **absolutely specific** detection of gases via the low pressure rotational fingerprint of gases in the THz. The new elements which make this interesting are (1) solid state source developments (currently market driven and mass produced at ~100 GHz and realizable but not yet to mass market ones to ~500 GHz), and (2) a conceptual development which replaces the traditional slow and complex phase lock loop technology with a fast scan and digitization scheme (see the description of the FASSST system in the attached article).

The second is a **remote detector**. It is based on the modulation of the target gas via a double resonance scheme with a CO₂ TEA laser pump and THz probe. While each of the elements in the proposed scheme can be calculated, the parameter space in which it would operate is many orders of magnitude away from any experimental base and this scheme should be considered 'high-risk'. The double resonance plays two roles: (1) to provide a modulation to the target signature which will allow its separation from the much larger variations associated with atmospheric fluctuations, and (2) much higher specificity by adding the pump frequency (the laser is tunable and the ir pump absorptions are a function of frequency) and the molecular relaxation signatures to the THz absorption spectrum signature. The resulting signature space is 3 dimensional and thus contains many more elements.

A. Point Detection

The Doppler limited spectra (linewidths of ~ 1 MHz) of gases in the THz is *absolutely specific*, even in relatively complex mixtures. Additionally, because the interactions between THz radiation and molecules are so strong in the submillimeter, such a point detector can be very sensitive. This can be seen from the discussion above and is discussed in more detail in the attached article from *Analytical Chemistry*.

Briefly put, the proposed scheme is to develop compact (a few in³), low power consumption (3 V), and potentially very low cost (\$100 - \$1000) versions of the BWO based system that was the basis for the article in *Analytical Chemistry*.

Figure 8 shows a 'brass board' version of such a system. In this prototype system, the molecular absorption cell would be milled into a solid block of metal of dimensions ~10 cm x 10 cm x 1 cm. By meandering, the cell could have an absorption path length of ~ 1 m. A small pump would reduce the pressure to ~0.1 Torr and standard preconcentration schemes to remove the atmospheric oxygen, nitrogen, and perhaps water could be used to increase the sensitivity at some cost in size and complexity. The detector output could either be digitized and analyzed on board, or directly telemetered (a 1 MHz rate would be standard, but rates as low as ~10 kHz are probably viable) to a central location.

The source and detector modules are much smaller than the cells and would be attached directly to the input and output of the molecular absorption cells. Rather than the conventional phase lock methodology, the gas detector would operate as the FASSST system described in the attached. In this scheme frequency calibration (to $1/10^5$ or $1/10^6$) is done in software, based on the approximately known ($\sim 1\%$) frequency/voltage characteristic of the source and calibration from known spectral lines (in some applications a second cell for the calibrating gas would be desirable).

With current commercial components, the low-end version would be based on an ~ 100 GHz source that has been developed either for the collision avoidance or wireless communications markets. A mid-level system based on current components would be based either on sources developed in the 200 - 500 GHz region for the radio astronomy and atmospheric communities or a broad band (~ 100 - 600 GHz) photomixer source developed in parallel at Lincoln Laboratory. The bandwidth of the latter would make it a particularly attractive source. The mid-level system would have capabilities similar to those described in the attached article, while the frequency restrictions of the low-end system would reduce its generality and sensitivity.

B. Remote Detection

The challenges associated with spectroscopic remote detection are much greater than with point detection primarily because of the much greater linewidth (~ 5000 MHz) associated with atmospheric pressure broadening. This greatly reduces the specificity of spectroscopic techniques in all spectral regions. At least as importantly, it makes the small absorptions associated with trace concentrations difficult to separate from atmospheric fluctuations (It should be noted that for concentrations defined as fractional partial pressures (e. g. ppm), the absorption strength in the laboratory at low pressure and the field at atmospheric pressure are the same).

Figure 9 shows a double resonance scheme based on a CO_2 TEA laser pump and THz probe. In this scheme (which is in effect a very long, very high pressure Optically Pumped Far Infrared Laser) the pump and probe are copropagated through the atmosphere. If the TEA laser has enough power to saturate the pump transition (which for the two test cases for which there is data at low pressure and high pressure calculation have been made (CH_3Cl and CH_3F) they do), the THz absorption/emission is **modulated** on the time scale of the atmospheric molecular relaxation ($\sim 10^{-10}$ s). This modulation, whose amplitude is the same order of magnitude (or perhaps larger) than the trace gas absorption, is on a time scale which should be easy to separate from atmospheric fluctuations and other clutter. Although I know little about radar signal processing, it would appear that there is a great similarity between this signal processing problem and those associated with short pulse radar. Although an active probe based on backscatter is shown, passive THz detection may be equally favorable, especially for sky viewing geometries.

One of the interesting features of this scheme is that its specificity is three-dimensional. In addition to the dimension associated with the THz probe frequency, there is another associated with the infrared pump absorption frequency (the TEA laser is tunable) and a third associated with the molecule specific time scale of the molecular relaxation which connects the ir pump pulse to the THz probe. **Figure 10** shows an example. In this case the CO_2 TEA laser pump would modulate the CH_3Cl signature near 266 GHz, but not the much stronger HCN which is nearly coincident in the THz spectrum.

Finally, as pointed out by Tim Grayson, there is a possibility that the atmospheric fluctuations can be overcome by observing simultaneously (or on a time scale shorter than the atmospheric fluctuations) enough of the atmospheric signature to determine the fluctuations in water concentration, temperature, and pressure. Then these contributions could be subtracted from the channels containing the target gas signatures. In comparison to the proposed double resonance scheme this has the advantage of much greater simplicity and covertness, but would depend on a model of the atmosphere which has a better understanding of the continuum contributions to absorption in the windows than we currently have. To a spectroscopist who likes to study such things and believes that it is possible to build such a model, this is not an overwhelming negative! Although there are many unknowns in both approaches, I would guess that the double resonance system would offer considerably higher specificity.

Some Gases of Interest at 1 ppm Dilution

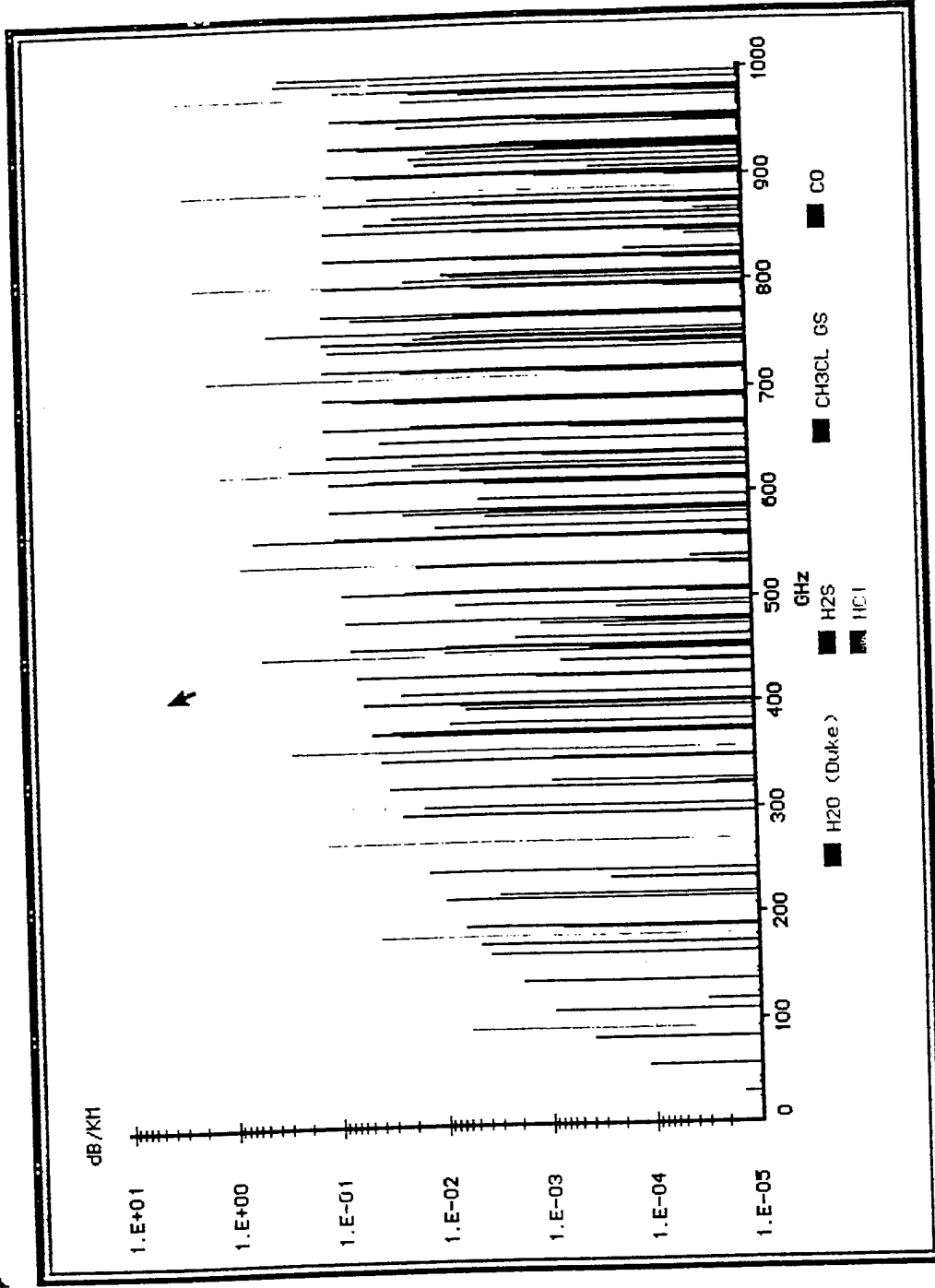


Figure 1. Absorption coefficients of some common gases at 1 ppm dilution.

SO₂ at 1 ppm Dilution 100 GHz Region

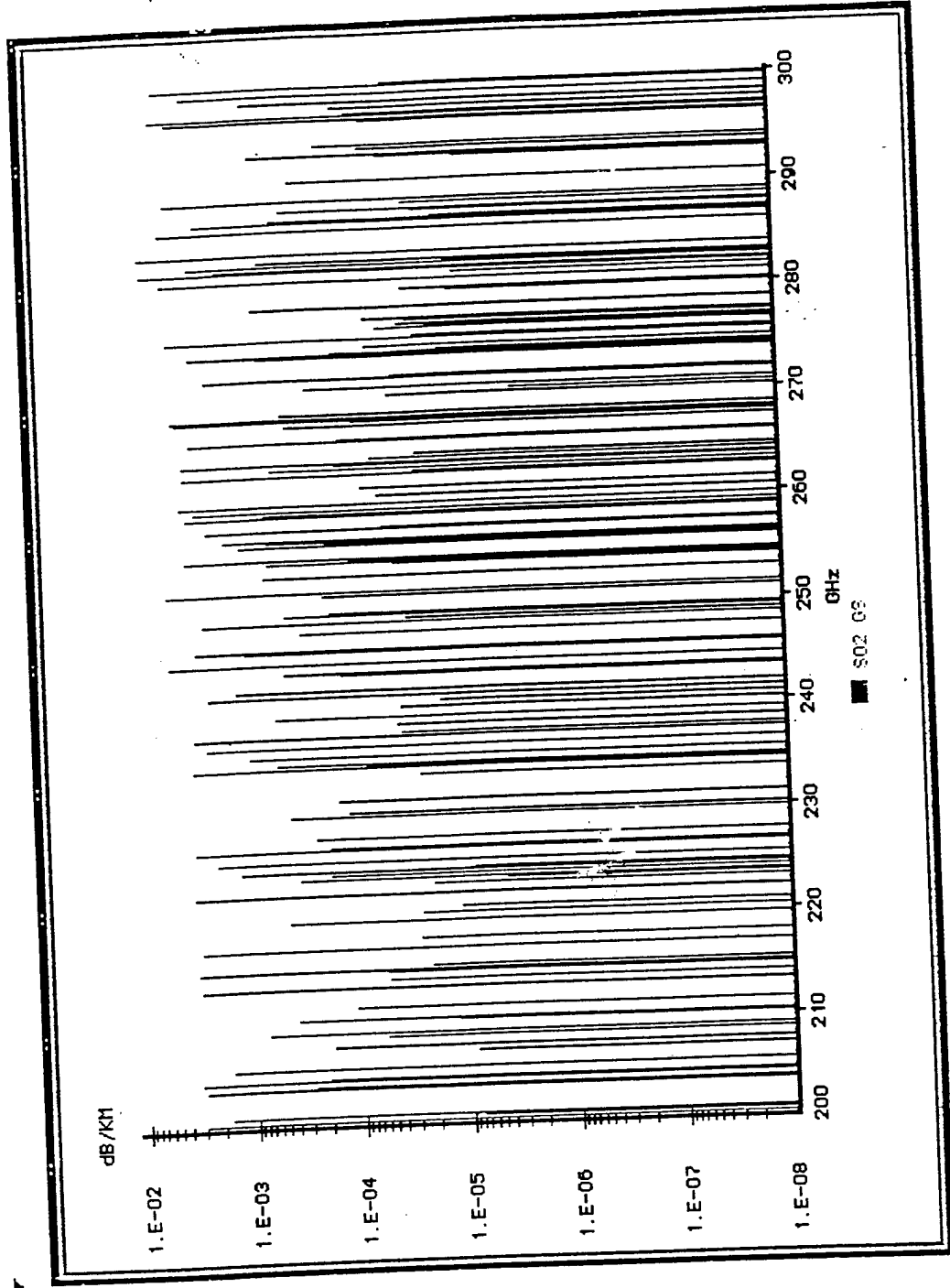


Figure 2. Spectrum of SO₂ between 200 and 300 GHz at 1 ppm concentration.

SO₂ at 1ppm Dilution 1 GHz Region

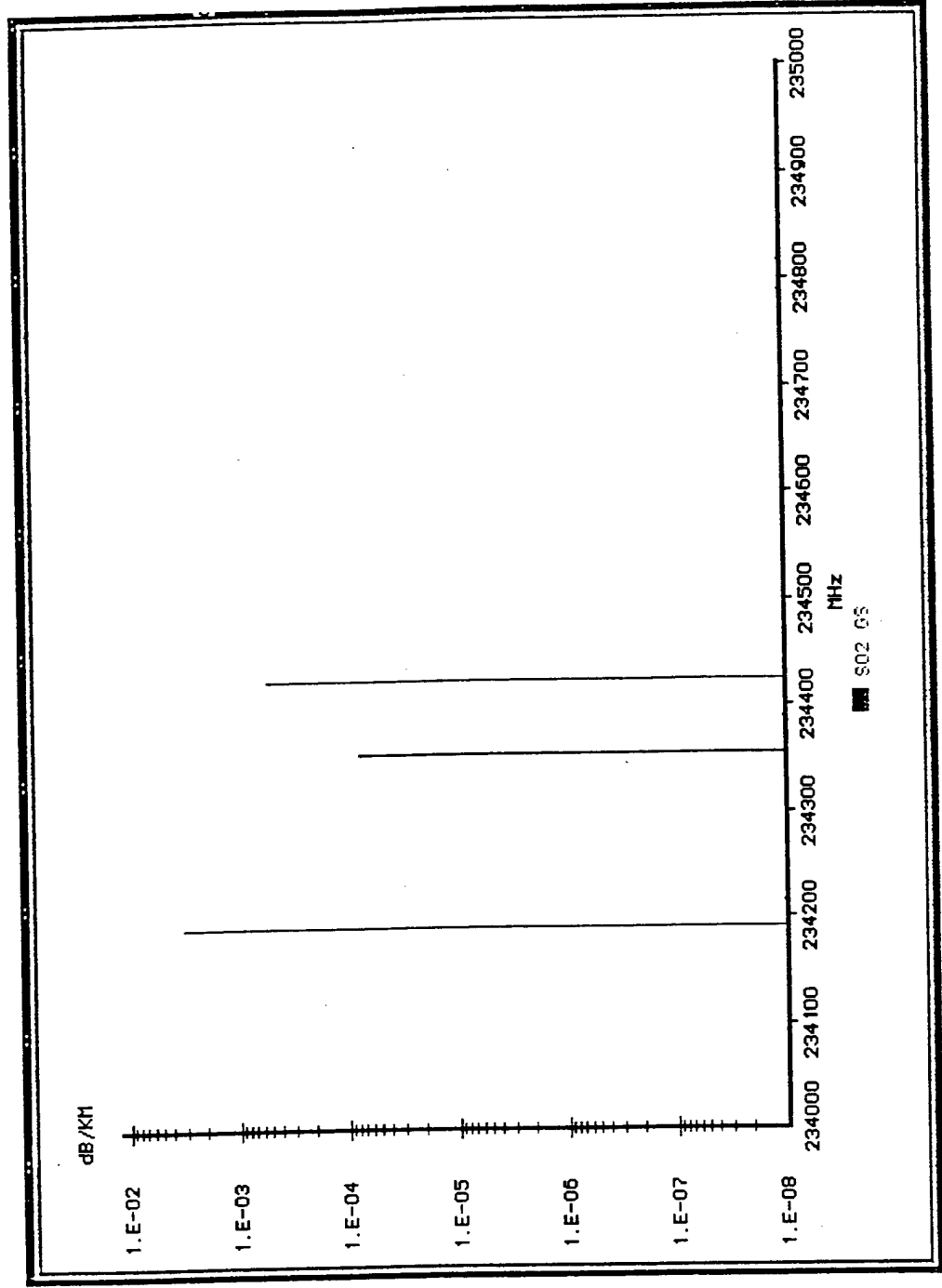


Figure 3. A 1 GHz region of the spectrum of SO₂ at 1 ppm dilution. At Doppler resolution the linewidths would be $\sim 1/5$ of the plotter line width.

Some Gases of Interest at 1 ppm Dilution 100 GHz Region

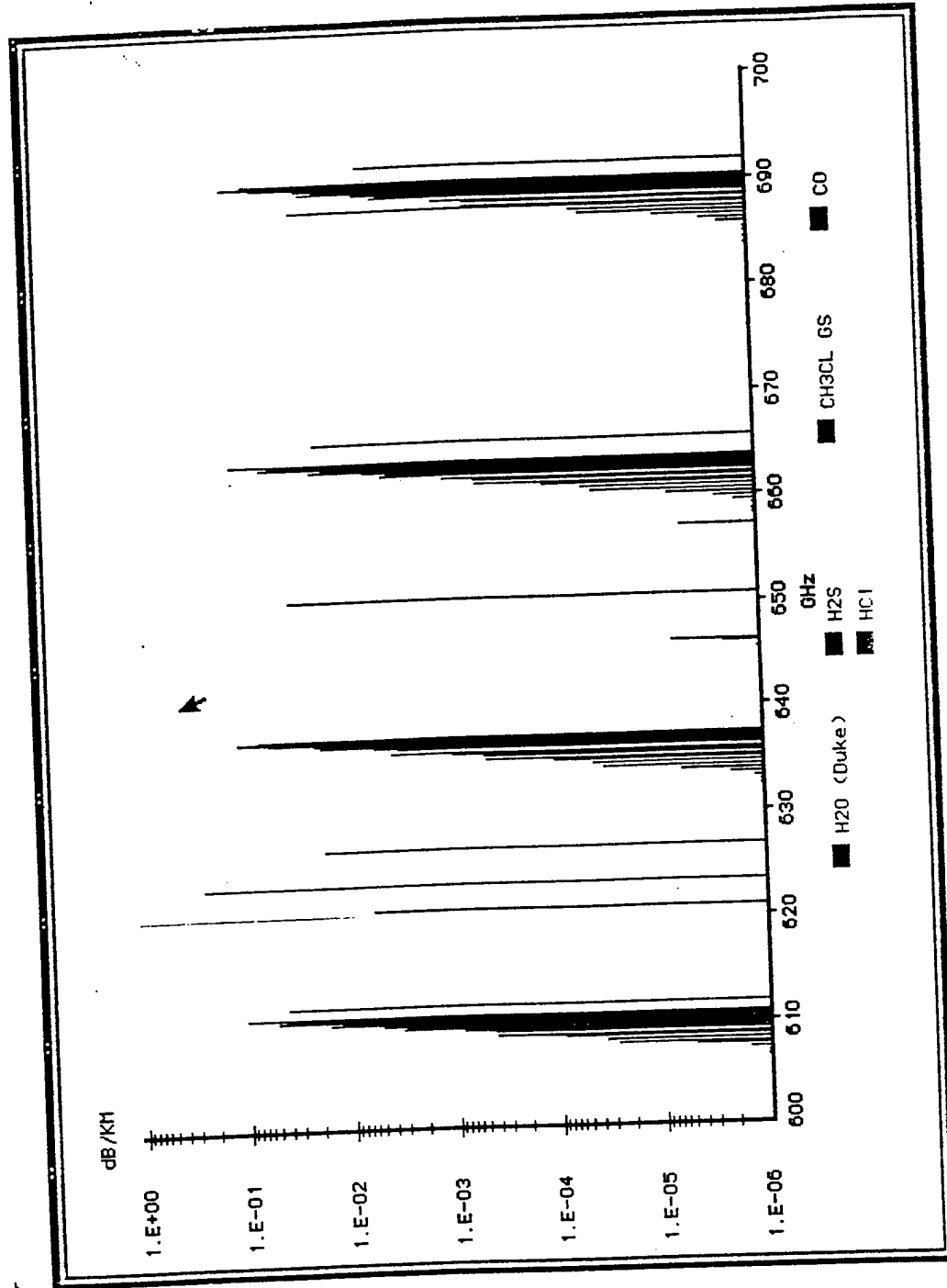
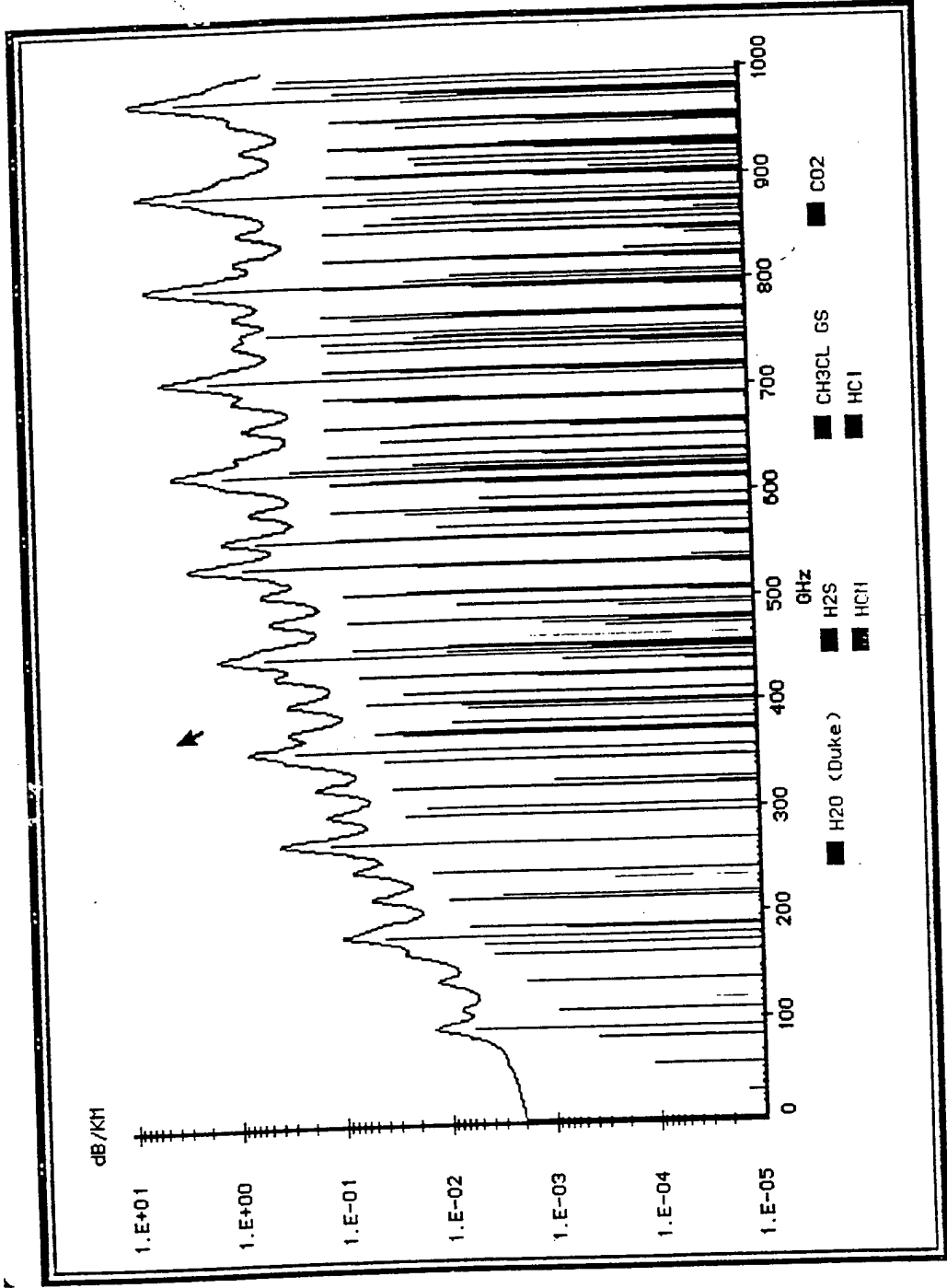


Figure 4. A 100 GHz region of Figure 1 showing in more detail some of the spectral character.

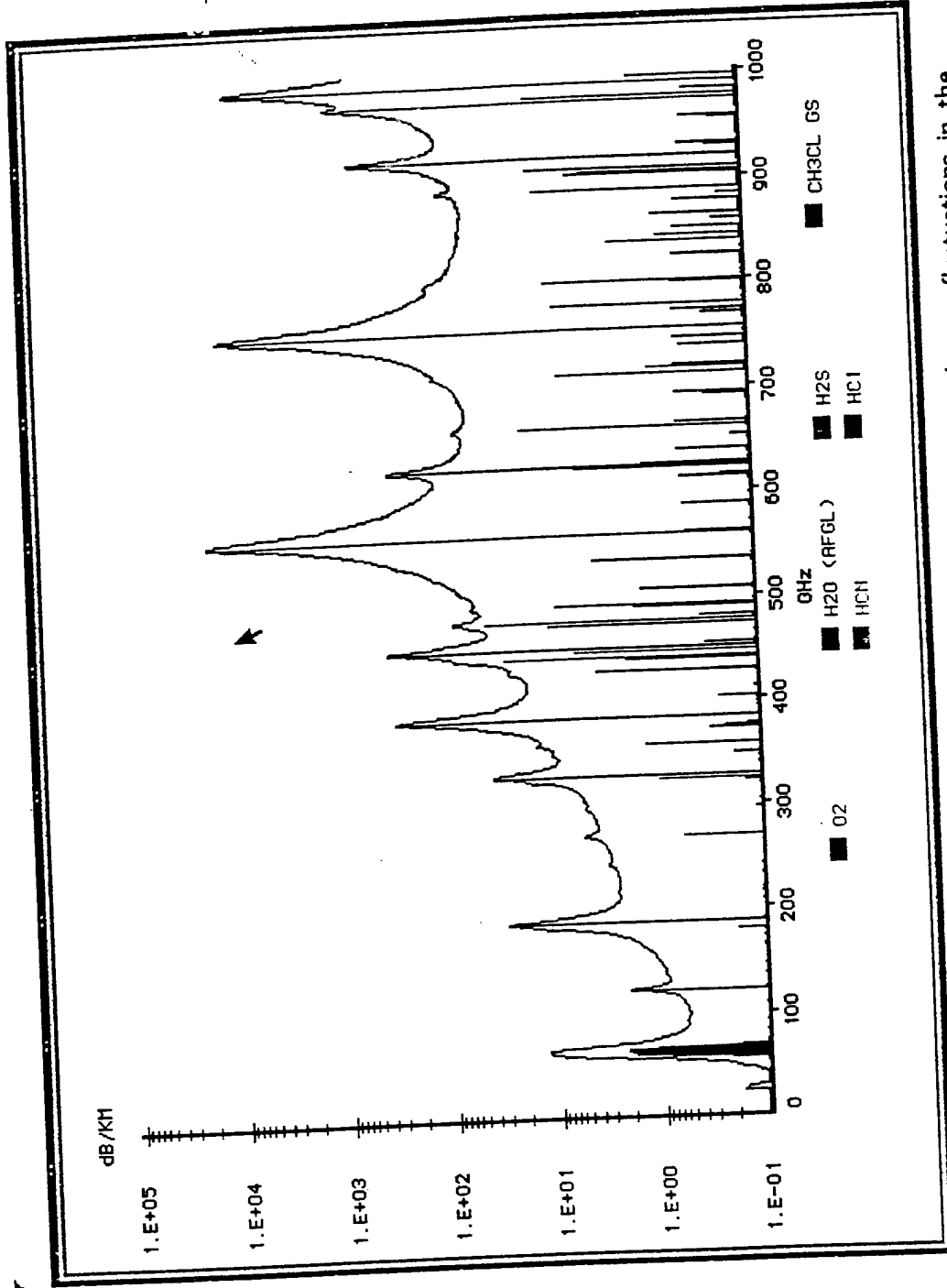
Some Gases of Interest at 1 ppm Dilution Broadened to Atmospheric Width



For active system with 1 second integration, noise $\sim 1.E-06$

Figure 5. The spectrum of Figure 1 broadened to atmospheric width.

Some Gases of Interest at 1 ppm Dilution in the Real Atmosphere

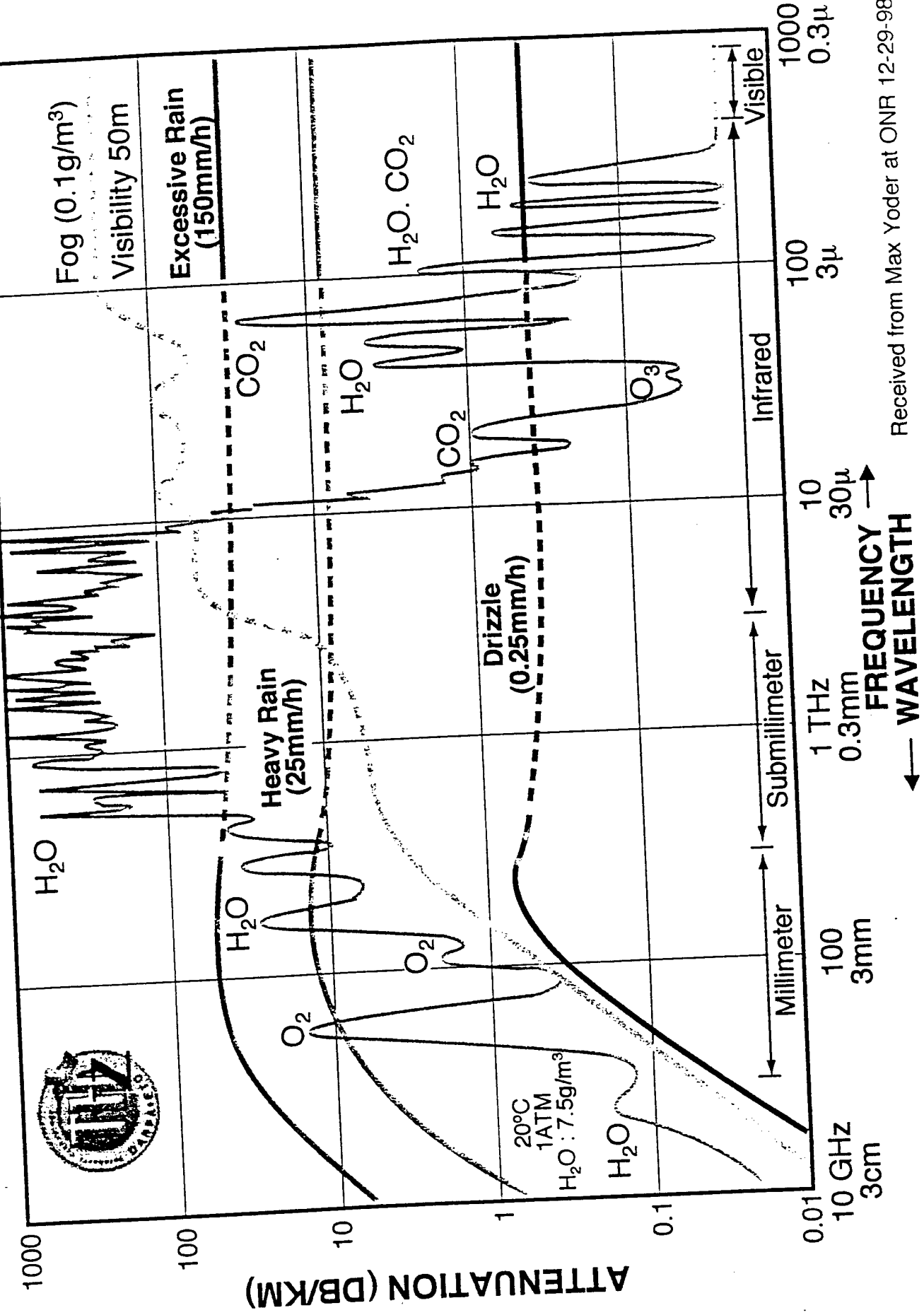


For an active or passive system, noise level will be determined by the unknown fluctuations in the Atmosphere's THz transmission

A 'high-risk' solution to this problem will be proposed below

Figure 6. A real atmospheric spectrum added to the trace gases of Figure 5.

E/M Attenuation vs Frequency



Received from Max Yoder at ONR 12-29-98

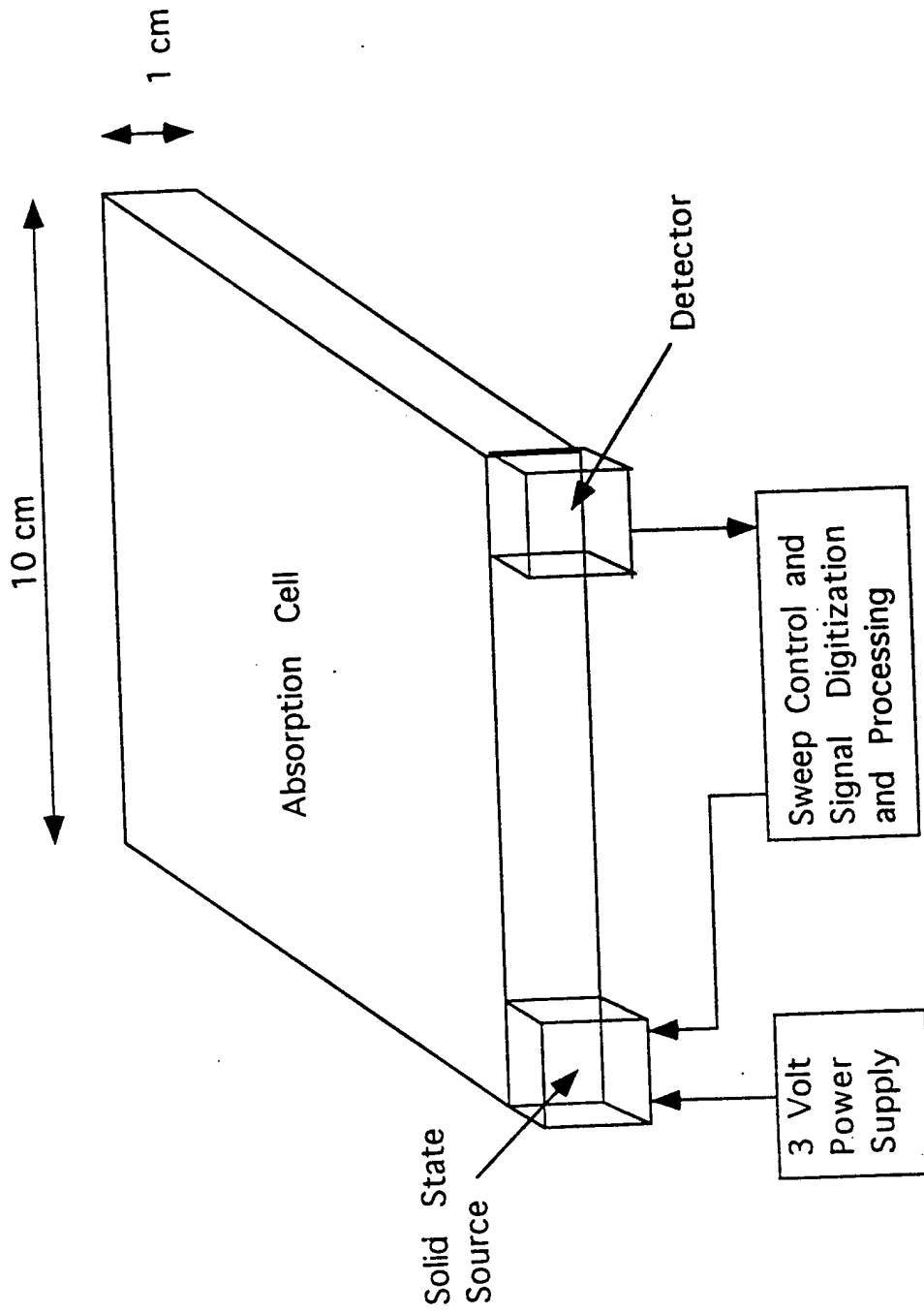


Figure 8. Point detector based on solid state source and detector technology and FASSST THz system concept.

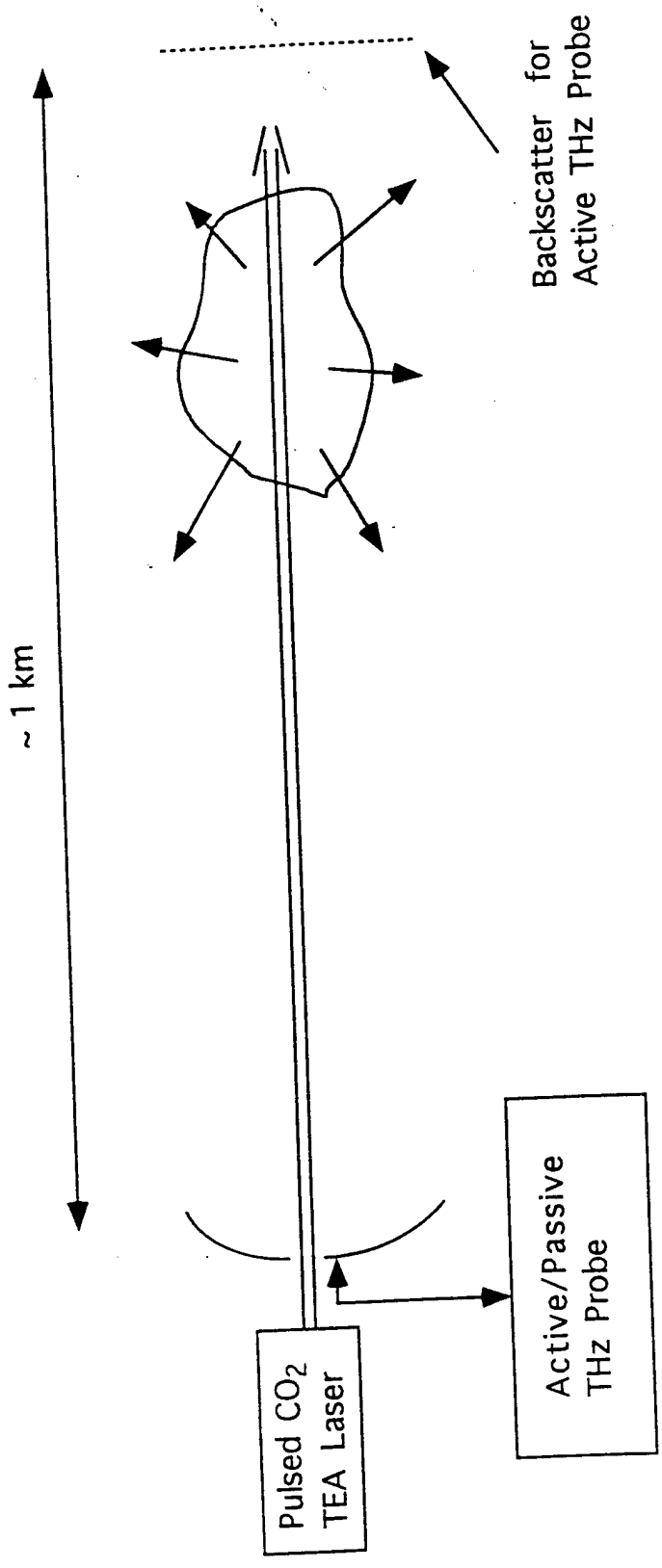


Figure 9. Geometry for remote detection of trace atmospheric gases based on THz/IR double resonance.

Pump specificity for CH₃Cl against HCN which is not vibrationally in resonance with the infrared pump

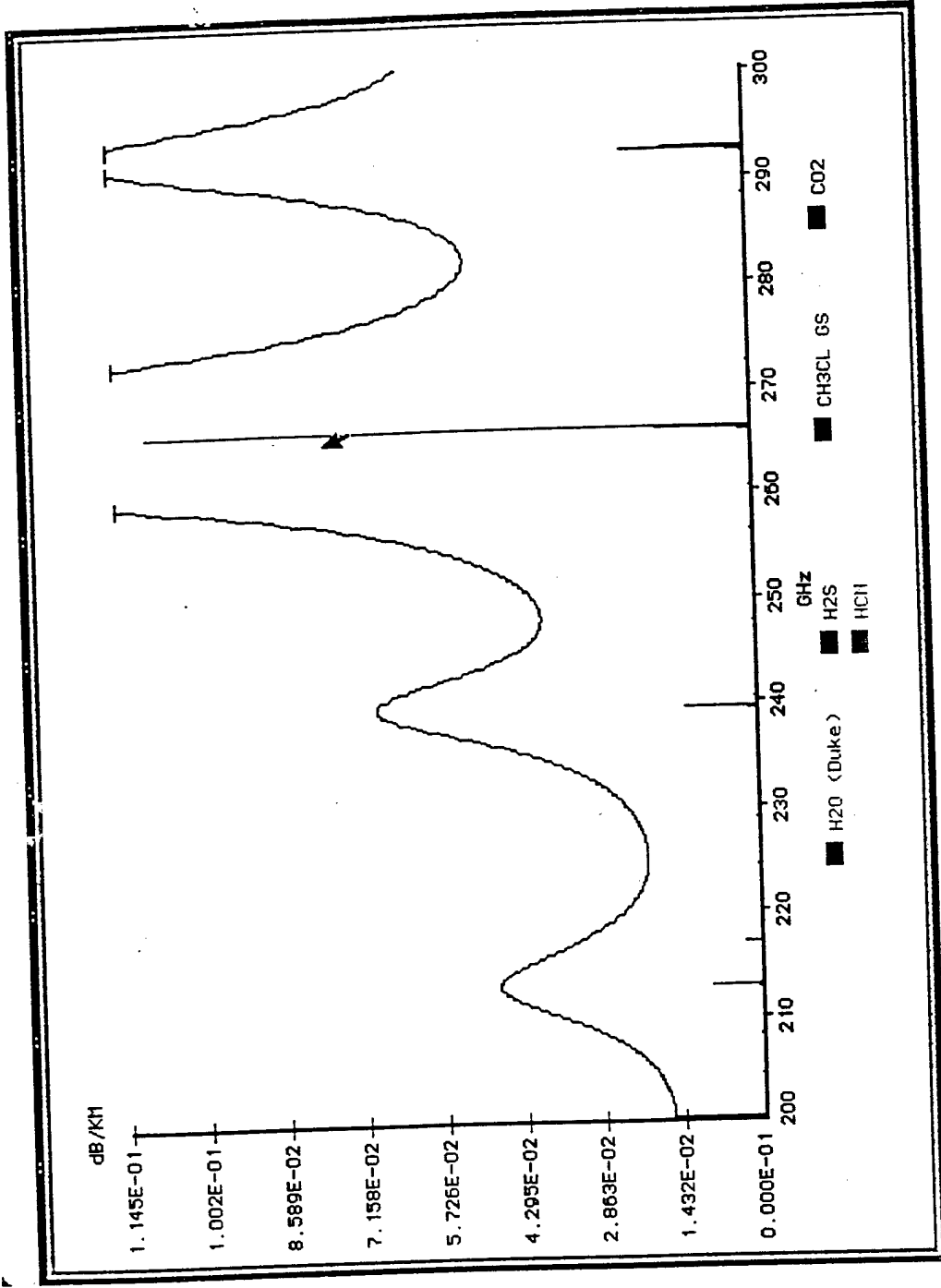


Figure 10. Portion of the spectrum of Figure 1 which shows the coincidence between an HCN and CH₃Cl line which would be removed by the lack of a similar pump coincidence in the infrared.