United States
Environmental Protection
Agency

Research and Development

Health Effects
Research Laboratory
Research Triangle Park NC 27711

EPA/600/S1-87/002 Aug. 1987

EPA

Project Summary

Millimeter-Wave Emissivity of Cellular Systems

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A general analysis is presented of the millimeter-wave and far-infrared spectroscopic properties of *in vivo* cellular systems, and of the boson radiative equilibrium with steady-state nonequilibrium molecular systems. The frequency threshhold of spectroscopic properties associated with such nonequilibrium effects is (roughly) estimated to be as low as ~100 GHz, if the Fröhlich vibrational model can be invoked for cellular-membrane systems. On this basis, the rationale for the specific experimental protocol employed was further to utilize the fact that in a photosynthetic preparation the onset and disappearance of the nonequilibrium state can be switched by modulating the optical-illumination input. The negative findings of the present V-band (50-75 GHz) radiometric study on *in vivo chlorella pyrenoidosa* algal cells appear to confirm the absence of non-Planckian emissivity features in the total radiometric study on *saccharomyces cerevisiae* near 42 GHz *(3)*, but does not negate totally the existence of nonequilibrium emissivity terms in the frequency range studied. Limitations of radiometer sensitivity, in spite of the S/N enhancement achieved through the AM-modulation technique, and/or damping of the non-Planckian emissivity features may have vitiated their detection. More fundamentally, the frequency threshhold for such properties may indeed lie at higher frequencies, in the far-infrared region. An order of magnitude of refinement in the theoretical models will be required to serve as a reliable guide for the prediction of such frequency threshholds.

This Project Summary was developed by EPA's Health Effects Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Discussion

As a consequence of the existence of nonequilibrium thermodynamic subsystems in *in vivo* biological entities, including cellular systems, spectroscopic features may be expected to arise which differ from those exhibited by ordinary equilibrium thermodynamic systems. A detailed theoretical analysis identifies, in particular, the emissivity function as a *targetable observable in this category*. The emissivity function, $\epsilon(\omega)$, is defined as the ratio of the actual emission intensity of the system under consideration, at a given frequency, to that of an ideal blackbody at the same temperature. The emission intensity is the photon energy emitted across *a unit surface area, per* unit time, into a solid angle of 2π steradians, and is proportional to the value of the generalized distribution function of photons over frequencies, $n(\omega)$, at the given frequency, characteristic of the thermodynamic system under consideration. If the system is an ideal blackbody, $n(\omega)$ is the Planck function and $\epsilon(\omega)$ is unity and frequency-independent.

A general thermodynamic analysis shows that nonequilibrium systems should exhibit frequency-dependent features in $\epsilon(\omega)$. If the Froehlich vibrational model is applied to cellular systems and the cellular membrane is identified as the locus of nonequilibrium thermodynamic processes, a frequency regime as low as ~ 100 GHz is credible for the possible existence of non-Planckian features. This study utilized *in vivo* photosynthetic algal cells [*chlorella pyrenoidosa* (American Type Culture Collection, in the form of their strains ATCC 22521, 7516 and 11469)] as the experimental model system, since the photosynthetic apparatus is located in the cell membrane, and since photosynthetic processes lend themselves directly to external modulation. In consequence, the temporal onset and disappearance of these nonequilibrium processes are externally controllable, and the opportunity is provided to improve the S/N ratio of measurements of the contribution of the nonequilibrium subsystem(s) to the total emissivity function, the preponderant portion of which arises from the Planckian background from the remainder of the system.

Millimeter-wave radiometry systems usually operate within the frequency regions encompassed by individual wavequide systems. This study employed V-band [WR-15], 50-75 GHz, to roughly approximate the frequency domain charted in the application of the Froehlich model to the photosynthetic chloroplast. The latter consists of a flat, double-membraned configuration, with a membrane thickness between 100-200 Å, the site of the photosynthetic apparatus and the putative locus of longitudinal vibrations invoked in the model. A serious *caveat* in any generalization that might be made from the results of this type of study arises from the combination of the crudeness of the frequency-range estimate from theory and the inherent narrow-bandedness of microwave and millimeter-wave waveguide systems.

The emissivity of the cellular preparation was measured using a 50-75 GHz radiometer, with its detection circuit phase-locked to the optical illumination of the sample in the wavelength range centered at 4900 Å (±350 Å), AM-modulated by a precision optical chopper at 100 Hz, to accomodate the response time constant of the photosynthetic apparatus of the preparation, and to provide selectivity and S/N ratio enhancement of the radiometer signal arising from processes associated with the photosynthetic apparatus. The source of the optical illumination was a 1000W Xe lamp, followed by standard components of an optical train, including a (distilled-H_2O) broadband infrared filter and optical filters selected to tailor the illumination to the wavelength response function of the cellular preparation. Millimeter-wave emission from the sample was measured at an angle of 90° to the optical-illumination axis, with the cell preparation contained in a quartz cuvette, transparent to both visible light and millimeter-wave emission. In the context of the present experiments, the task of the radiometer was to detect any response, phase-locked to the optical illumination, within a primary-illumination range of 200-1000W total (regulated and calibrated) power, of the cell preparation.

The local-oscillator (LO) arm of the radiometer originated in a set of three klystron oscillators spanning the 50-75 GHz range, and was coupled to the sample arm and the detection-system arm via broadband (50-75 GHz) ferrite isolators. The frequency axis of the radiometer signal was provided by measurement of the LO frequency by means of a millimeter-wave electronic counter and subsequent conversion of its frequency output via a digital/analog (D/A) converter to a voltage analog signal to comprise the Y-axis of an X/Y recorder output. Under typical experimental conditions, frequency resolution thus obtained is equal to or better than 0.1 MHz, over the entire V-band frequency range, and frequency tracking of the LO is automatic. A separate power-measurement loop permits determination of the LO relative power across a given klystron mode, as a function of LO frequency. The primary element of the radiometer detection system is a broadband (50-75 GHz) harmonic mixer matched to an intermediate-frequency (IF) amplifier, the latter rejecting IF signals below ~ 500 MHz and above ~ 1500 MHz. Electronic tuning of the LO, by means of its reflector voltage, moves the center of this IF detection window (bandwidth ~ 1000 MHz) across the klystron mode. Director monitoring of radiometer performance is accomplished by feeding the output of the IF amplifier directly into a (0-2 GHz) spectrum analyzer. Radiometry of the sample preparation is achieved by entering the output of the IF amplifier into a microwave video detector, thus, converting that output into a dc voltage. The phase-lock synchronization loop, externally synchronized to the precision optical chopper, is engaged via a phase-sensitive lock-in amplifier which accepts the dc radiometer-output signal. Finally, the radiometer-signal axis, from the output of the lock-in amplifier, is combined with the frequency axis, from the electronic counter and D/A converter, to present the radiometry data on an X/Y recorder.

The present study failed to detect any (non-artifactual) frequency-dependent features in the radiometry signal phase-locked to the optical illumination of the photosynthetic algal-cell preparation. Among the possible explanations for such a finding, two are salient. The central modality of the radiometer experiment (direct AM-modulation of the photosynthetic process) may have been vitiated by insufficiently favorable S/N ratio characteristics of any non-Planckian features in the emissivity. This would render the sensitivity of the current radiometer design incapable of detecting such features, in a manner which excludes artifacts of millimeter-wave waveguide circuits. From a mechanistic standpoint, any of three (heuristic) parameters may have contributed to such a possibility. (a) the low concentration of subsystems which undergo nonequilibrium processes, per unit volume, compared to the (bulk) system near equilibrium; (b) low absolute emission intensity associated with any non-Planckian features; and/or (c) damping of any non-Planckian emissivity features, with an attendant reduction in S/N ratio, within a given frequency interval.

The second case arises from the central question, to which existing theory is only a very rough guide, concerning the low-frequency threshhold, ω_1, of non-Planckian features. If $(n\ \omega_1/\kappa T) \ll 1$ is inadmissible as an assumption for cellular systems, no non-Planckian features are expected in the frequency range studied, with T≅300°K, and their absence in the present study is not inconsistent with the thermodynamic or the generalized Froehlich model, but would imply that definitive refinement in the theory is required for the prediction of their (frequency) range of existence.

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The complete report, entitled "Millimeter-Wave Emissivity of Cellular Systems," (Order No. PB 87-175 840; Cost: $11.95, subject to change) will be available only from:
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EPA/600/S1-87/002