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TECHNICAL PAPER proposed for presentation at National Heat Transfer Conference sponsored by the American Society of Mechanical Engineers Seattle, Washington, August 6-10, 1967

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ABSTRACT

The validity of the assumption that local thermodynamic equilibrium is present in a gas is examined with respect to its effect on the calculated emission of thermal radiation. The spectral distribution of energy emitted by the gas is determined when there is departure from equilibrium. Attention is confined to the case of insufficient interparticle collisions being available to redistribute the energy of absorbed radiation. Thus the equilibrium distribution of energy states in the gas cannot be maintained. Emission of radiation from such a gas is not in the usual spectral energy distribution determined by the equilibrium spectral absorption coefficient and the Planck distribution.

Specifically, the case of the grey gas is used to derive some approximate relations for characterizing the conditions where departure from equilibrium might be expected.

INTRODUCTION

Solutions to radiative energy transfer problems involving absorbing-emitting media almost invariably contain the assumption of "local thermodynamic equilibrium" (LTE). Basically this means that any small gas volume is assumed to be in thermodynamic equilibrium so that its temperature can be defined.

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When this assumption is valid, it greatly simplifies radiant transfer problems. The emission of energy from a volume element can be specified in terms of the Planck spectral energy distribution and the radiative absorption coefficient regardless of the spectrum of the flux passing through the element. If LTE cannot be assumed, the Planck distribution is not valid and the actual distribution, along with the gas absorption coefficient, becomes a function of the population of the energy states in the gas. Detailed quantum-statistical calculations become necessary to describe accurately the emission and absorption of radiant energy. In the general case involving surface-gas and gas-gas radiant energy exchange, consideration of such detailed phenomena tax present analytical methods past their capabilities.

It is possible to envision physical systems where nonequilibrium effects could be significant. Some of these have been examined by Kulander [1] and Oxenius [2]. For example, if very large temperature gradients are present such as those in shocks or those expected in proposed gaseous-core nuclear rockets, then species migrating to a given position may come from regions at widely differing conditions. One temperature could hardly be expected to characterize local conditions in such a case. Ferrari and Clark [3,4] have examined this situation with reference to hypersonic shock phenomena.

A less common example is the presence of transients in the incident radiation. If the transients are quite rapid so that significant changes in incident radiation intensity occur over periods of time of the same order as those governing the emission process in the gas (electron transitions, vibrational transitions, etc.), then the emission of energy may greatly lag the absorption process. This is a nonequilibrium situation and might be encountered in connection with thermal emission from a nuclear fireball.

Another case is that of a rarefied gas exposed to a strong external radiation field. Here the gas particles are excited to high energy states by the absorption of radiant energy. If sufficient collisions could occur, this energy would be distributed into an equilibrium distribution of energy among the possible states by numerous particle interactions, and the Planck distribution and gas absorption coefficient would govern the emission of radiation. However, in the rarefied gas, insufficient collisions may occur. Emission of energy may then be in a distinctly non-Planckian distribution because energy is radiated by spontaneous transitions from these nonequilibrium excited states.

This paper is concerned with the latter case. An approximate grey-gas analysis is carried out to delineate roughly the range of variables that determines significant departure from LTE.

ANALYSIS OF NONEQUILIBRIUM RADIATION FROM A GREY GAS

Consider a volume element dV of grey gas with linear absorption coefficient α, exposed to radiation from a blackbody source at a temperature T_s. What is desired is a representation of the spectral distribution of intensity emitted by dV if LTE is not assumed.

If steady state is considered, then the total energy emitted by dV per unit time, Q_e, must be equal to the energy absorbed by dV per unit time, Q_a. If dV were in thermodynamic equilibrium, the conservation of radiative energy would be expressed by

$$Q_a = Q_e = \alpha(F\sigma T_s^4)dV = 4\alpha\sigma T_{eq}^4\ dV \tag{1}$$

where F is the geometric configuration factor between the source and dV, and T_{eq} is the equilibrium temperature of the gas. The equilibrium temperature of the gas can be related to F and T_s by equation (1) as

$$T_{eq} = \left(\frac{F}{4}\right)^{1/4} T_s \tag{2}$$

If the energy leaving dV per unit time is not in an equilibrium spectral distribution, total energy must still be conserved. The total energy leaving dV per unit time is then given by

$$Q_{e,ne} = 4\pi\alpha\ dV \int_0^\infty i_{\lambda,ne}\ d\lambda \tag{3a}$$

Since total energy is conserved, $Q_{e,ne}$ must still equal Q_a, which by equation (1) is

$$Q_{e,ne} = 4\pi\alpha\ dV \int_0^\infty i_{\lambda,ne}\ d\lambda = \alpha\left(F\sigma T_s^4\right) dV \tag{3b}$$

An accurate representation of $i_{\lambda,ne}$ can only be obtained by detailed analysis of the population of the various energy states in the gas, and the various transition processes between these states and their probability of occurrence. However, by some judicious assumptions, it is possible to get approximate results.

Consider emission from dV, as an approximation, to be composed of two parts. Each part corresponds to equilibrium spectral emission based on a given temperature, and these parts will be combined to give the nonequilibrium spectral distribution.

The first part is energy that is absorbed from the incident beam, and is then emitted spontaneously by the gas <u>before</u> being distributed by interparticle collisions. This part is assumed to have a spectral distribution similar to that from the blackbody source at T_s, which is a Planck distribution given by $i_{\lambda,eq}(T_s)$. Such an assumption is valid if the spontaneous emission causes transition of the particle to the original unexcited state. Then energy absorbed at a given wavelength will be spontaneously reemitted at the same wavelength unless a collision occurs prior to such emission. This criterion of return to the original state is exactly satisfied in the two-level atom, which is often used for studies of transition processes, but becomes increasingly poor as more and more energy states become possible in the gas being considered [5].

The remaining fraction of the absorbed energy is assumed to be emitted in a spectral distribution $i_{\lambda,eq}(T_{eq})$ corresponding to the equilibrium temperature T_{eq} of dV. This emission is assumed to be from that portion of dV that remains in an equilibrium state. Then the intensity at a given wavelength is

$$i_{\lambda,ne} = A' i_{\lambda,eq}(T_s) + A i_{\lambda,eq}(T_{eq}) \tag{4}$$

This synthesis of two equilibrium spectral distributions is analogous to that occurring when scattering processes in gray gases are studied.

Integrating equation (4) over all wavelengths and substituting equation (3b) gives

$$\int_0^\infty i_{\lambda,ne}\, d\lambda = \frac{F\sigma T_s^4}{4\pi} = \frac{A'\sigma T_s^4}{\pi} + \frac{A\sigma T_{eq}^4}{\pi} \tag{5}$$

Solving for A' gives, using equation (2)

$$A' = \frac{F}{A} - A\left(\frac{T_{eq}}{T_s}\right)^4 = (1 - A)\left(\frac{T_{eq}}{T_s}\right)^4 \tag{6}$$

Equation (2) was derived for LTE. It is valid, however, to use it in the non-LTE case, equation (6), because equations (1) and (2) for the relation between T_s, F, and T_{eq} must also hold for that portion of dV that is assumed to be in equilibrium so long as only a gray gas is considered. This is clear from examination of equation (1): If some fraction of Q_a is absorbed by the equilibrium portion of dV, then the same fraction of Q_e in equation (1) must be emitted by the equilibrium portion of dV. Equation (2) thus holds for the portion of dV assumed to be in thermodynamic equilibrium at temperature T_{eq}. Finally, equation (4) can be written

$$i_{\lambda,ne} = (1 - A)\left(\frac{T_{eq}}{T_s}\right)^4 i_{\lambda,eq}(T_s) + Ai_{\lambda,eq}(T_{eq}) \tag{7}$$

This equation provides the form of $i_{\lambda,ne}$ for the model assumed. To obtain quantitative results, the value of the parameter A must be estimated. This parameter can be interpreted as the probability of a given radiatively excited gas particle returning to the equilibrium population based on T_{eq} by means of an interparticle collision before undergoing a spontaneous radiative emission from the excited state.

Suppose that there are N_o excited particles at some given time. If it is assumed that some number N_R of these excited particles emit spontaneously in some time interval t, then assuming a first order rate process,

$$dN_R = \frac{N}{\bar{\tau}_R} dt \tag{8}$$

where $\bar{\tau}_R$ is the mean lifetime of a radiatively excited state, and N is the remaining excited population at time t. Similarly, if N_c of the particles undergo collisions in time t,

$$dN_c = \frac{N}{\bar{\tau}_c}\,dt \tag{9}$$

where $\bar{\tau}_c$ is the mean time between collisions.

Equations (8) and (9) can be integrated to obtain N_R and N_c at some time t. The probability that the particle deexcites by means of a collision is

$$A = \frac{N_c}{N_R + N_c} = \frac{\dfrac{1}{\bar{\tau}_c}\displaystyle\int_0^t N\,dt}{\dfrac{1}{\bar{\tau}_R}\displaystyle\int_0^t N\,dt + \dfrac{1}{\bar{\tau}_c}\displaystyle\int_0^t N\,dt} = \frac{\bar{\tau}_R}{\bar{\tau}_R + \bar{\tau}_c} \tag{10}$$

Substituting this result into equation (7) gives

$$i_{\lambda,ne} = \frac{1}{\left(\dfrac{\bar{\tau}_R}{\bar{\tau}_c}\right) + 1}\left[\left(\frac{T_{eq}}{T_s}\right)^4 i_{\lambda,eq}(T_s) + \frac{\bar{\tau}_R}{\bar{\tau}_c}\, i_{\lambda,eq}(T_{eq})\right] \tag{11}$$

Equation (11) can also be placed in the dimensionless form

$$\frac{i_{\lambda,ne}}{i_{\lambda,eq}(T_{eq})} = \frac{1}{\left(\dfrac{\bar{\tau}_R}{\bar{\tau}_c}\right) + 1}\left\{\left[\left(\frac{i_{\lambda,eq}(T_s)}{T_s^5}\right)\Bigg/\left(\frac{i_{\lambda,eq}(T_{eq})}{T_{eq}^5}\right)\right]\frac{T_s}{T_{eq}} + \frac{\bar{\tau}_R}{\bar{\tau}_c}\right\} \tag{12}$$

This form has some utility because it illustrates the departure from equilibrium in the emitted spectral distribution of a gas, and is in terms of the tabulated blackbody functions $\left[i_{\lambda,eq}(T)/T^5\right]$ (ref. [6]) and the two parameters $(\bar{\tau}_R/\bar{\tau}_c)$ and (T_s/T_{eq}).

DISCUSSION OF NONEQUILIBRIUM GREY GAS EFFECTS

Effect of ratio of transition to collision times. - Examination of equation (11) shows that the intensity of emitted radiation from a grey gas as a function of wavelength is very dependent on the ratio of $\bar{\tau}_R$, the mean lifetime of an excited state before a radiative transition, to $\bar{\tau}_C$, the mean time between interparticle collisions. As $\bar{\tau}_R/\bar{\tau}_C$ goes to zero, meaning that many radiative transitions occur before a collision can take place, the emitted spectrum approaches that of the incident radiation reduced in magnitude by the factor $(T_{eq}/T_s)^4$. As $\bar{\tau}_R/\bar{\tau}_C$ approaches large values, only the last term on the right of equation (11) is significant, and the emitted spectrum approaches that of an equilibrium gas.

Equation (11) has the same form as that for the intensity leaving a gas with combined absorption and isotropic coherent scattering. For that case $\bar{\tau}_R$ and $\bar{\tau}_C$ are replaced by the absorption and scattering coefficients, respectively [7].

In figure 1, the spectral distribution of intensity from a gas volume when $(T_{eq}/T_s) = 0.1$ is plotted for various values of $\bar{\tau}_R/\bar{\tau}_C$, assuming $T_s = 10\ 000^\circ$ K. Although the distortion due to the log-log scale must be considered, it is obvious that even for $\bar{\tau}_R/\bar{\tau}_C = 10$, the spectrum is quite different from the equilibrium Planck spectrum that is present when $\bar{\tau}_R/\bar{\tau}_C \to \infty$.

The departure from equilibrium is also illustrated in figure 2, again for $(T_{eg}/T_s) = 0.1$. Here, the ratio of $i_{\lambda,ne}$ to $i_{\lambda,eq}(T_{eq})$ is shown as a function of λT_{eq}. For $\bar{\tau}_R/\bar{\tau}_C \to 0$, which is essentially the case of no collisions in the gas being available for the redistribution of energy, the largest deviation from equilibrium is seen to occur. Emission is in the same spectral distribution as the absorbed energy.

It is interesting that all values of the parameter $\overline{\tau}_R/\overline{\tau}_C$ give an intensity ratio of unity at the one λT_{eq} value of 1500 μm $^{\circ}$K for this case. Thus, if this analysis were completely valid, it would be impossible to discern nonequilibrium effects by examining the emitted energy from the gas at this λT_{eq}, where T_{eq} would be found in a practical calculation from equation (2). Conversely, this is the only λT_{eq} value at which the equilibrium temperature of the gas could be determined by external observation because no nonequilibrium effects distort the emitted intensity.

Also shown on figure 2 is a dotted curve indicating the fraction of equilibrium intensity that lies below a given λT_{eq} value. This can be used to determine the importance of the departure from equilibrium at various λT_{eq} values. For example, the curve for $\overline{\tau}_R/\overline{\tau}_C = 10$ deviates by less than 10 percent from the equilibrium value for λT_{eq} values greater than 1500 μm $^{\circ}$K. Over 98 percent of the equilibrium intensity lies in this range. Therefore, even though the deviation of $[i_{\lambda,ne}/i_{\lambda,eq}(T_{eq})]$ from equilibrium reaches factors as high as 100 at λT_{eq} of about 800, the intensity of equilibrium radiation at these λT_{eq} values is so small that the spectrum of intensity from the gas remains within 10 percent of equilibrium in all important spectral regions.

For $\overline{\tau}_R/\overline{\tau}_C \rightarrow 0$, however, deviations from equilibrium by factors of 10^{-3} occur over λT_{eq} ranges containing significant values of intensity.

Effect of source temperature. - In the general problems of interest to engineers, T_{eq} cannot be specified but rather is defined by equation (2). In the discussion presented here, it has been assumed that the interchange factor F is of such magnitude that it gives the ratios of (T_{eq}/T_s) used in the examples. Equations (11) and (12) could be rewritten to eliminate either T_s or T_{eq} in terms of the interchange factor, but the present formulation seems most useful. In figure 3, the intensity ratio is plotted as a function of the source-to-equilibrium gas temperature ratio with $\bar{\tau}_R/\bar{\tau}_C$ as a parameter. This is done for two values of the remaining parameter λT_{eq}. It is obvious that the deviation from equilibrium is always greater as $\bar{\tau}_R/\bar{\tau}_C$ becomes smaller, regardless of the source-to-gas temperature ratio. The effect of (T_s/T_{eq}), however, depends upon the specific λT_{eq} value being observed.

At $(T_s/T_{eq}) = 1$, that is for isothermal conditions, all curves pass through $i_{\lambda,ne}/i_{\lambda,eq}(T_{eq}) = 1$ so that, as expected, equilibrium is present for isothermal conditions regardless of λT or $(\bar{\tau}_R/\bar{\tau}_C)$.

<u>Parameters governing the departure from equilibrium</u>. - To determine readily when the analysis used here predicts significant departure from LTE, some additional computations can be made for simple cases. Let $i_{\lambda,eq}(T_{eq})$ be given by Planck's distribution

$$i_{\lambda,eq}(T_{eq}) = \frac{2C_1}{\lambda^5[\exp(C_2/\lambda T_{eq}) - 1]} \approx \frac{2C_1}{\lambda^5 \exp(C_2/\lambda T_{eq})}$$

where the final approximate form, known as Wien's distribution, is within 1 percent of Planck's distribution for λT less than 3000 $\mu m\ ^{\circ}K$. Substituting this final form into equation (13) yields

$$\frac{i_{\lambda,ne}}{i_{\lambda,eq}(T_{eq})} = \frac{\psi(\lambda) + (\bar{\tau}_R/\bar{\tau}_C)}{1 + (\bar{\tau}_R/\bar{\tau}_C)} \tag{13}$$

where

$$\psi(\lambda) = \left(\frac{T_{eq}}{T_s}\right)^4 \exp\left[\frac{C_2}{\lambda T_{eq}}\left(1 - \frac{T_{eq}}{T_s}\right)\right] \tag{14}$$

Use of Wien's distribution should not introduce significant error into equation (14) even at $\lambda T_{eq} > 3000\ \mu m\ ^{\circ}K$, because it appears as a ratio.

For equilibrium to be present, the left-hand side of equation (13) should be unity. Examination of equation (13) for this case shows that equilibrium is present when either of the conditions

$$\psi(\lambda) \approx 1 \tag{15}$$

or

$$\frac{\bar{\tau}_R}{\bar{\tau}_C} \gg 1 \quad \underline{\text{and}} \quad \frac{\bar{\tau}_R}{\bar{\tau}_C} \gg \psi(\lambda) \tag{16}$$

are met. Condition (15) predicts equilibrium for source and equilibrium gas temperatures nearly equal while condition (16) shows that for $\bar{\tau}_R$ sufficiently greater than $\bar{\tau}_C$, other parameters become unimportant.

The λT_{eq} value at which equilibrium always is present for a given (T_{eq}/T_s) is found by substituting equation (14) into equation (15) and solving for λT to obtain

$$(\lambda T_{eq})_{eq} = \frac{C_2\left(1 - \frac{T_{eq}}{T_s}\right)}{4 \ln\left(\frac{T_s}{T_{eq}}\right)} \tag{17}$$

<u>Departure from equilibrium in gases under assumed conditions</u>. - For the curves of figures 1 to 3, it appears that $\bar{\tau}_R/\bar{\tau}_C$ of 10^2 or greater would assure a close approach to equilibrium energy emission from the grey gas for the case of T_s/T_{eq} of 10. To examine what this means in terms of temperature and pressure, a representative case may be examined.

Consider a grey gas that attains a temperature of 300° K when exposed to a source of radiation at an effective blackbody temperature of 3000° K. First let us see if either of the conditions given by equations (15) and/or (16) are satisfied. The value of $\psi(\lambda)$ is, using $C_2 = 14388\ \mu m\ ^\circ K$,

$$\psi(\lambda) = 10^{-4} \exp\left(\frac{C_2}{\lambda T_{eq}} \cdot 0.9\right) = 10^{-4} \exp\left(\frac{1.294\times10^4}{\lambda T_{eq}}\right) \tag{18}$$

Examination of figure 2 shows that less than 2 percent of the emitted equilibrium energy is at $\lambda T_{eq} < 1.5\times10^3$, while more than 98 percent lies at $\lambda T_{eq} < 2\times10^4$. For these two λT_{eq} values, $\psi(\lambda)$ has the values of 0.562 and 1.91×10^{-4}, respectively. For $\bar{\tau}_R/\bar{\tau}_C = 10^2$, the condition of equation (16) is certainly met. What conditions in the gas will give a transition time to collision time ratio of this magnitude?

The value for mean time between collisions in a gas composed of hard spheres is approximated from the kinetic theory of gases to be, for a gas in equilibrium,

$$\bar{\tau}_C = \frac{1}{N\bar{\sigma}_C\bar{v}_R} \tag{19}$$

where N is the number of particles per unit volume, $\bar{\sigma}_C$ is the mean collision cross section, and $\bar{v}_R$ the mean relative velocity of two particles. If the number of particles at standard conditions is given by Loschmidt's number, $Lo = 2.687\times10^{19}$ atoms per cubic centimeter, the N is given by

$$N = Lo \cdot \frac{P}{P_o} \cdot \frac{T_o}{T} = 7.34\ \frac{P}{T} \times10^{21}\ \text{atoms/cm}^3 \tag{20}$$

where P is in atmospheres.

The value of $\bar{v}_R$ is also given by kinetic theory [8] as

$$\bar{v}_R = \sqrt{2}\bar{v} = \sqrt{\frac{6kT}{M}} \qquad (21)$$

where $\bar{v}$ is the mean velocity of the gas particles, k is Boltzmann's constant, and M the mass of the individual particles. This can be rewritten as

$$\bar{v}_R = 2.24\sqrt{\frac{T}{M}} \times 10^4 \text{ cm/sec} \qquad (22)$$

where m is the molecular weight of the gas. Substituting equations (22) and (20) into (19) gives

$$\bar{\tau}_C = \frac{\sqrt{mT}}{1.64\ \bar{\sigma}_C P} \times 10^{-26} \text{sec} \qquad (23)$$

If the gas under consideration were monatomic and of unity atomic weight then $\bar{\sigma}_C \approx 3 \times 10^{-15}$ cm^2 and m = 1. This provides an upper limit for the predicted nonequilibrium effects. For $T = 300^\circ$ K, equation (23) becomes

$$\bar{\tau}_C = \frac{3.53 \times 10^{-11}}{P} \text{ sec}$$

For monatomic gases, $\bar{\tau}_R$ is about 10^{-8} seconds [9], so that

$$\frac{\bar{\tau}_R}{\bar{\tau}_C} = 10^2 \approx \frac{P \times 10^{-8}}{3.53 \times 10^{-11}}$$

or

$$P \approx 0.35 \text{ atm}$$

Thus, for a pure monatomic gas at 300° K exposed to radiation from a blackbody source at 3000° K, deviation from equilibrium might begin at as high a pressure as 1/3 atmosphere, becoming more severe as pressure is decreased.

For a molecular gas, lifetimes in a given vibrational or rotational energy state are on the average much longer than the lifetimes of excited electronic energy states in monatomic gases. Penner [9], for example, quotes molecular energy state lifetimes as being of the order of 10^{-1} to 10^{-3} seconds. Collision times for molecules are not significantly different from atoms, however, because both m and $\bar{\sigma}_C$ increase for molecules, but appear as a ratio in equation (23). Therefore, for a molecular gas, deviations from equilibrium will not occur until pressures perhaps 10^{-6} times smaller than for an atomic gas. Such conditions are not often encountered except in studies of radiative interaction with interstellar gases.

CLOSING REMARKS

The model used in deriving equation (12) has some serious shortcomings for quantitative use. Primarily, of course, it is derived for a grey gas, a highly unrealistic situation of interest chiefly as an aid to understanding. Further, it was assumed that the gas absorption coefficient was unaffected by nonequilibrium considerations; in a real gas, this is not true. The latter assumption and those outlined in the analysis, however, are not greatly in error when the departure from equilibrium is small. Thus, the analysis has some utility in predicting the parameter values which signal the departure from equilibrium. The equations (15) and (16) thus may give an indication of when departure from equilibrium radiative emission might occur.

LIST OF SYMBOLS

A', A — emission probabilities defined by equation (4)

C_1, C_2 — constants in the Planck spectral energy distribution

F — geometric interchange factor

i — intensity; energy per unit projected area per unit solid angle per unit time

$i(\lambda)$	spectral distribution of intensity; intensity per unit wavelength
k	Boltzmann's constant
Lo	Loschmidt's number; 2.687×10^{19} atoms/cm^3 at STP
M	mass of particle
m	molecular weight of particle
N	number of particles per unit volume
P	pressure, atm
Q	energy per unit time
$\bar{v}$	mean velocity
$\bar{v}_R$	mean relative velocity
T	absolute temperature
t	time
α	linear radiation absorption coefficient
λ	wavelength
σ	Stefan-Boltzmann constant
$\bar{\sigma}_C$	mean collision cross section
$\bar{\tau}_C$	mean time between collision of excited particle with another particle
$\bar{\tau}_R$	mean lifetime of excited state before radiative spontaneous transition
$\psi(\lambda)$	function defined by equation (14)

Subscripts:

a	absorbed
c	collisional deexcitation
e	emitted
eq	at equilibrium conditions
ne	at nonequilibrium conditions

o at standard conditions

R radiative deexcitation

s at source of blackbody radiation

0 at time $t = 0$

λ spectrally dependent

E-3789

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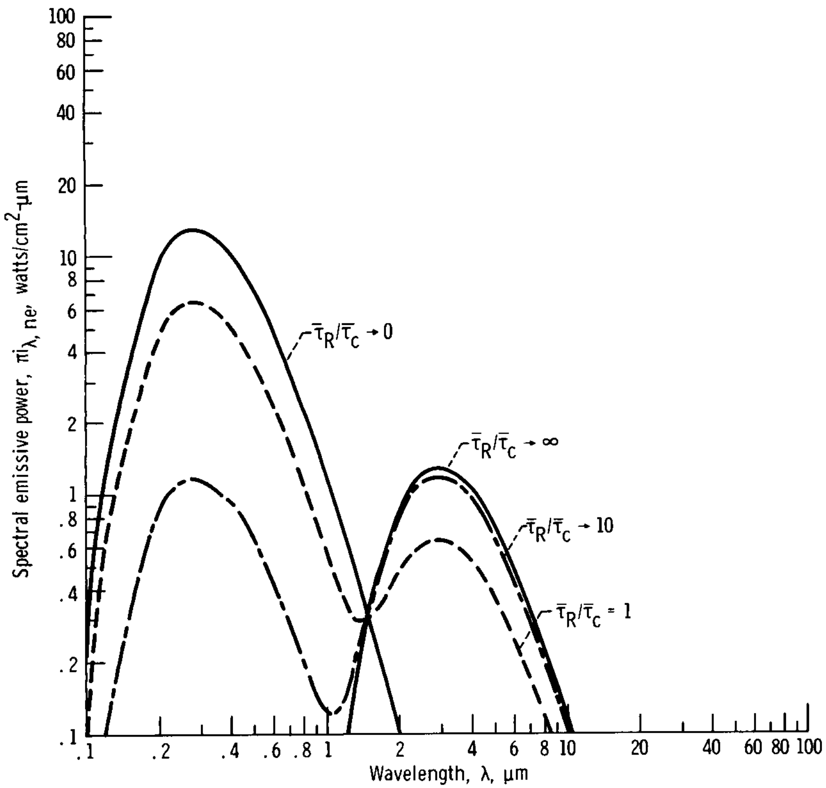

Fig. 1. - Emission spectrum from nonequilibrium grey gas. Source temperature, $T_s = 10^4$ °K, $(T_{eq}/T_s) = 0.1$.

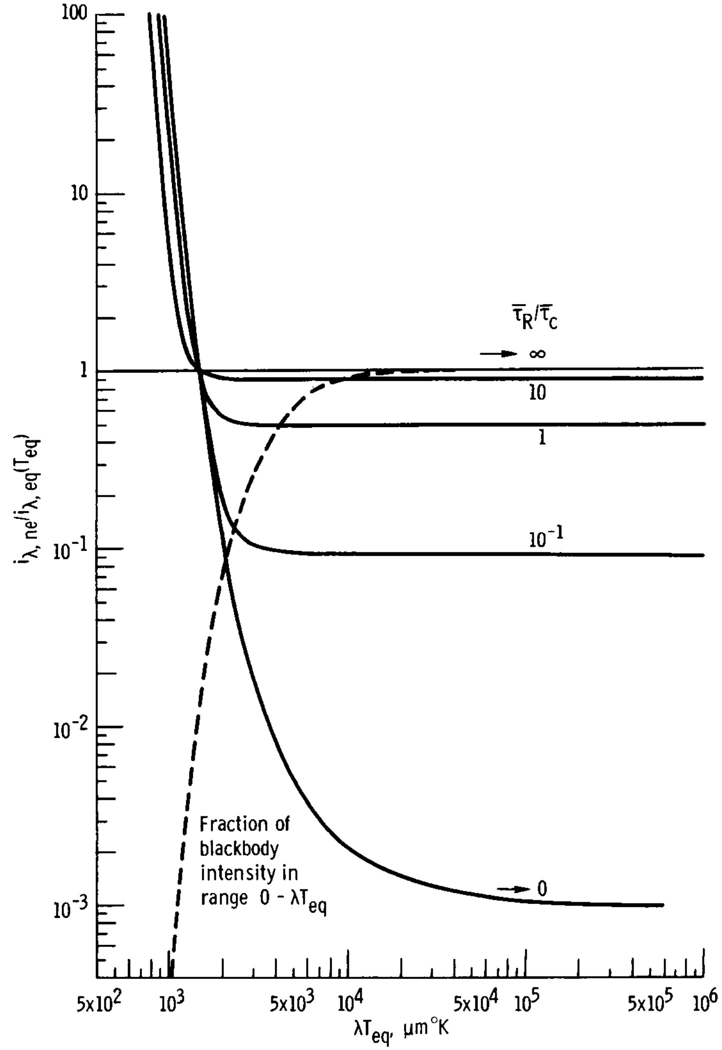

Fig. 2. - Departure from equilibrium of a grey gas as a function of radiative transition time to collision time; $T_s/T_{eq} = 10$.

E-3789

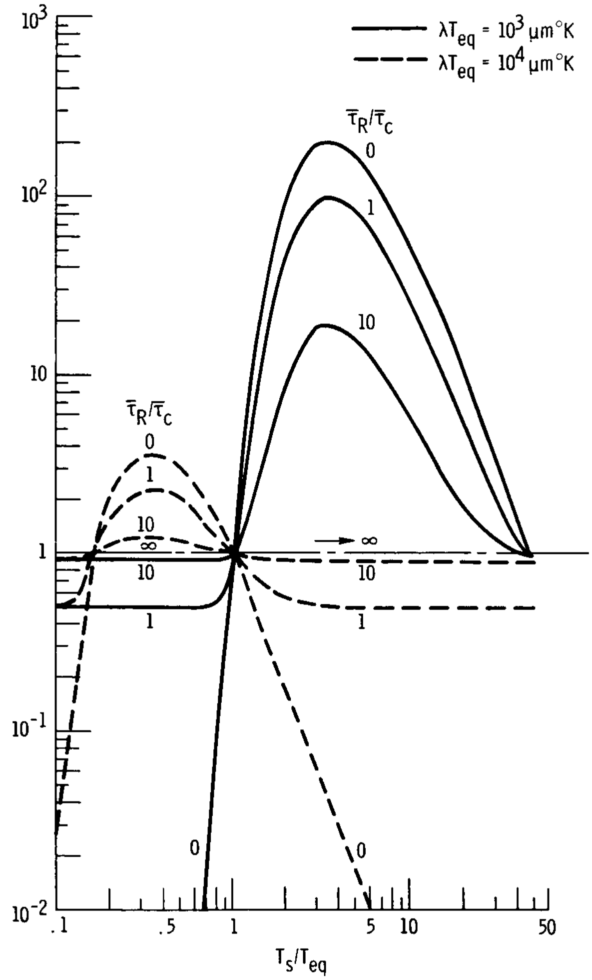

Fig. 3. - Effect of source temperature on emission from a nonequilibrium gas.