

# CONVERSION OF LASER ENERGY TO GAS KINETIC ENERGY\*

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INTRODUCTION

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The use of a high intensity laser as a source of power can be advantageous in situations where it is either necessary or desirable to separate the sites of energy production and consumption by large distances. Such situations clearly arise in space applications where, for purposes of minimizing weight loading, it would be valuable to be able to supply various rocket or satellite power needs from Earth-based sites. Specific systems of this sort, which are presently under investigation, include (1) the novel idea of supplying rocket thrust via laser propulsion (ref. 1) and (2) direct Earth-based powering of satellites or space laboratories.

The successful development of such systems requires, among other things, efficient techniques for conversion of the incident laser radiation into other more useful forms of energy. This discussion is concerned with the direct gas phase absorption of laser radiation. The process envisioned is the heating of a high pressure reservoir gas via laser absorption followed by expansion of the heated gas through a sonic nozzle with subsequent conversion of the gas translational temperature to ordered kinetic energy. The resulting high velocity gas may then be used as specific impulse for rocket propulsion or alternately for the generation of shaft horsepower.

## GAS PHASE ABSORPTION MECHANISMS

### Inverse Bremsstrahlung

There are two basic techniques available for the gas phase absorption of laser radiation. The first of these is absorption by inverse bremsstrahlung. This technique is particularly suitable for absorption in high temperature gases and has been the subject of considerable research. The advantages of inverse bremsstrahlung are two fold: (1) the laser energy is converted directly to gas translational energy during the absorption process, that is, at a gas kinetic rate; and (2) the absorption process is spectrally continuous and thus lasers of any wavelength may be used.

The basic complication in the use of inverse bremsstrahlung is that the absorption coefficient scales either linearly or as the square of the electron density, depending on the ionization fraction, and thus under equilibrium conditions the absorption coefficient can be quite small at low temperatures. Even if a readily ionizable seed, such as cesium, is introduced into the gas, the absorption coefficient will still decrease exponentially with decreasing temperature below  $\approx 2500^\circ\text{K}$  and be negligibly small at room temperature. This complication can be avoided by

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producing non-equilibrium electron concentrations in the gas, for example, by laser induced gas breakdown or by an electric discharge; alternately, it can be avoided by introducing a different absorption mechanism that could be used to heat the gas to temperatures sufficiently high so that significant thermal ionization would occur.

### Molecular Absorption

The second gas phase absorption technique, and the principal subject of this discussion, is the direct absorption by the vibrational rotation (V/R) bands of molecules. The main advantage in using V/R band absorption is that, unlike bremsstrahlung, the absorption process is as efficient in a cold gas as in a heated one. However, there are several compensating disadvantages:

1. A given molecule will have a finite number of absorbing transitions which will occur over a limited wavelength range.
2. The absorbed energy is deposited in the vibrational rather than translational modes of the molecule and efficient kinetic mechanisms are required to convert the absorbed energy into heat. In particular, when the upper state of an absorbing transition is populated more rapidly than it can be de-activated, gas "bleaching" can occur.
3. As the gas temperature increases undesirable chemical phenomena, such as dissociation, can degrade the absorption process.

This last point is particularly important for larger molecules. For example, SF<sub>6</sub>, which is an efficient absorber of CO<sub>2</sub> laser radiation, will be significantly dissociated at temperatures above ~ 1700°K. Even a relatively stable triatomic such as CO<sub>2</sub> will dissociate at temperatures above ~ 2500°K. Generally speaking, only diatomic molecules with large dissociation energies can provide significant laser gas heating with minimal chemical effects. As will be discussed below, these disadvantages may be minimized by performing the absorption in a high pressure system. Although the discussion below will be limited to the absorption of diatomic molecules, much of what is said will also hold true for larger molecules.

### High Pressure Absorption

The pressure broadened absorption coefficient for a given V/R transition with vibrational levels  $v - 1 \rightarrow v$  and rotational level  $J \rightarrow J \pm 1$  is defined by

$$\alpha_{v-1, J, \Delta J}(\nu), \text{ cm}^{-1} = \frac{\lambda^2}{4\pi^2 c \Delta\nu_L} \frac{A_{v, v-1, J, \Delta J}}{N_{v-1, J} - \frac{g_{v-1, J}}{g_{v, J \pm 1}} N_{v, J \pm 1}} \left[ 4 \left( \frac{\nu - \nu_{v, J, \Delta J}}{\Delta\nu_L} \right)^2 + 1 \right]^{-1} \quad (1)$$

where  $\nu$  is wavenumber,  $c$  the speed of light,  $\lambda$  wavelength,  $A$  the Einstein coefficient of the transition,  $N$  the populations of the upper and lower states,  $g$  the degeneracy factor, and  $\Delta\nu_L$  the full linewidth at half height in  $\text{cm}^{-1}$ . It can be seen from the first bracketed term on the right-hand side of equation (1) that the absorption coefficient decreases as the upper state population approaches the lower. This is the phenomenon of "bleaching" alluded to earlier. The number density dependence of the absorption coefficient is of some interest. The line width is directly proportional to number density and thus on line center ( $\nu = \nu_{v,J,\Delta J}$ ) the absorption coefficient is independent of the total number density. On the other hand, on the wings of the line ( $\nu - \nu_{v,J,\Delta J} > \Delta\nu_L$ ) the absorption coefficient scales as the square of the number density. Thus there can be a large variation in the pressure dependence of the absorption coefficient over the line shape.

For any given vibrational transition there are a large number of allowable rotational transitions. For example, the room temperature absorption spectrum for the  $v = 0 \rightarrow 1$  transition of CO is shown in figure 1. It has been assumed here that the pressure is such that the line width is small compared to the spacing between lines ( $\sim 3.8 \text{ cm}^{-1}$  for CO). The  $v = 1 \rightarrow 2$  absorption spectrum will have a similar shape but will be shifted to somewhat longer wavelengths, as shown in figure 1, because of the molecular anharmonicity. As can be seen there is a considerable overlap between the  $v = 0 \rightarrow 1$  and  $v = 1 \rightarrow 2$  bands (and also with higher bands not shown); however if the line widths are small compared to the line spacing there will be essentially no wavelength coincidences between the various lines. Thus laser light of a given wavelength could at best be absorbed by only one of the manifold of possible absorbing transitions.

This constraint will not apply at sufficiently high pressures,  $\gtrsim 30 \text{ atm}$  for self-broadening in CO, where the line width is  $\gtrsim$  the line spacing. In this limit the molecule will absorb continuously over the wavelength region of the V/R band. The absorption coefficient at any given wavelength is no longer given directly by equation (1), but rather now involves a sum of the absorption coefficients of a number of overlapping transitions, and is independent of total number density depending only upon the mole fraction of the absorbing gas.

A band model has been developed for the prediction of the molecular absorption spectra in this high pressure limit and applied to the molecule CO. Predictions from this model are shown in figure 2 through 4. In figure 2 the room temperature absorption cross-section vs. wavelength is shown for the lowest three vibrational transitions of CO. The cross section increases with increasing vibrational level because of variation in the Einstein coefficient (a linear dipole moment function for CO was employed in the calculations (ref. 2)). The important feature is that, because of the large overlap between the absorption spectra for each vibrational transition, laser light of a given wavelength may be absorbed simultaneously by each vibrational level. This has the effect of minimizing bleaching phenomena because if a particular transition approaches the bleaching limit the upper state of that transition can also absorb the incident laser radiation.

Of more interest from the viewpoint of laser heating is the temperature variation of the absorption cross sections. This behavior will, of course, be quite sensitive to the choice of incident laser wavelength. Predictions for the absorption cross sections of the first five vibrational levels of CO over the temperature range of  $200\text{-}3000^\circ \text{K}$  are shown in figure 3. The incident laser radiation is taken to be at a wavelength of  $4.755 \mu$ , which would correspond to a P branch  $v = 1 \rightarrow 0, J = 10$  CO laser. Although a high power CO laser of this type has not yet been developed, the results may be taken as generic for any diatomic molecule undergoing absorption at a wavelength near the center

of its fundamental V/R band. As can be seen, above 300°K the absorption cross sections for the various vibrational levels are all of the same order of magnitude. The absorption cross sections for individual transitions generally tend to decrease at the higher temperatures shown because of the effect of the rotational partition function. However absorption on the higher vibrational transitions ( $V = 4 \rightarrow 5$  as well as higher transitions not shown) peak in the higher temperature region because absorption occurs in the high rotational wings of these bands. The net effect of this is that the overall molecular absorption coefficient can be relatively insensitive to temperature variations depending, of course, upon the vibrational distribution function.

A similar result is shown in figure 4 for an incident laser wavelength of  $4.855 \mu$ . This wavelength corresponds to an attainable P branch CO laser transition,  $V = 3 \rightarrow 2 \quad J = 8$ . Since this wavelength occurs in the wing of the CO  $V = 0 \rightarrow 1$  band (see figure 2), the absorption cross section for this initially increases rapidly with temperature. This is an important point since the upper vibrational states of CO are not significantly populated at room temperature. However, it is clear that as soon as ground state molecules are excited to the first vibrational state through absorption, excitation to higher states (including transitions to states higher than those shown in figure 4) will be quite rapid.

It would appear then that optimum absorption by diatomic molecules can be realized in the high pressure regime characterized by overlapping lines, since in this limit laser light of a given wavelength may be absorbed simultaneously on a number of different vibrational transitions. This multiline absorption should also have the effect of flattening out the temperature vibration of the total molecular absorption coefficient. It now remains to discuss techniques for the rapid conversion of the absorbed vibrational energy into translation.

#### Conversion of Vibrational Energy to Translation

The molecule CO was chosen as an example not only because its molecular properties are well defined, but also because it has a very high dissociation energy — 11.1 eV — and thus may be heated to relatively high temperatures ( $\sim 6000^\circ\text{K}$ ) without introducing significant chemical effects. One disadvantage of this molecule, however, is that the rate constants for vibrational deactivation are quite small at temperatures below  $1500^\circ\text{K}$ . These small rate constants tend to preclude efficient coupling between the vibrational and translational modes. There are several atomic species such as Fe, H, and O that have been shown to be efficient ( $k = 10^{-13}$  to  $10^{-12}$  cc/sec) vibrational deactivators of CO (refs. 3 through 5); however, it is difficult to introduce such species into a cold gas flow.

One technique has been considered that, although not recyclable, may be of some interest for use in systems concerned with the development of thrust, etc. The technique involves the use of a rare gas seeded with iron pentacarbonyl,  $\text{Fe}(\text{CO})_5$ .  $\text{Fe}(\text{CO})_5$  absorbs in the same wavelength region as CO, its vibrational modes will strongly couple with translation and, when dissociated, produces both the absorbing molecule CO and the efficient deactivator Fe. Unfortunately, detailed modeling of this system is complicated by the fact that the low temperature absorption spectra of  $\text{Fe}(\text{CO})_5$  is not well defined.

To properly model gas laser heating by diatomic molecular absorption one requires a computer code that includes the phenomena of multiline absorption, vibration to translational (V-T) deactivation, and vibration-vibration (V-V) exchange between all vibrational levels. The resulting

vibrational distributions will not only be out of equilibrium with translation but can also be non-Maxwellian. Preliminary calculations, neglecting the V-V exchange processes, have been made for CO. A constant density gas was assumed at an initial pressure of 30 atm with 0.2 percent CO, 0.04% Fe in a helium diluent. A laser intensity of  $10^6$  W/cm<sup>2</sup> was assumed with a laser wavelength of 4.855  $\mu$ , corresponding to the absorption cross sections of figure 4.

The time histories of both gas absorption length and temperature for this case are shown in figure 5. As can be seen, the gas temperature increases essentially linearly from 400° to 3000°K and, more importantly, the absorption length remains approximately constant over this temperature range. The initial drop in the absorption length is caused by the early time increase in the vibrational populations of the upper states. The inclusion of V-V exchange in the calculations is not expected to produce a large change in these predictions.

It should be emphasized that this technique may be generally applied. In general, the radiation from most high power gas lasers which operate on vibrational transitions can be absorbed by several different diatomic molecules if the total gas density is sufficiently high to produce overlapping lines. The one major exception to this statement appears to be the 10.6- $\mu$  CO<sub>2</sub> laser. Although a number of the diatomic metal oxides can absorb radiation of this wavelength, these species are not readily produced under the conditions of interest. Of course this same line broadening phenomenon occurs for larger molecules. These could be reviewed to determine which species could optimally absorb CO<sub>2</sub> laser radiation with minimal chemical effects.

There are a number of candidate diatomic molecules that can be vibrationally deactivated relatively efficiently and that may be of interest in a closed cycle laser heating system. For example, the room temperature absorption spectra of the first two vibrational levels of NO are shown in figure 6. The middle peaks in these spectra arise because the NO ground state is a  $\pi$  state and thus the Q branch is allowed. (For simplicity in this calculation a linear dipole moment was assumed and the small wavelength shift between spin states was neglected). Shown at the top of the figure are the wavelength positions of several readily attainable CO laser lines. It would appear that NO would prove to be a viable absorber for CO laser radiation.

## SUMMARY

Techniques for the gas phase absorption of laser radiation for ultimate conversion to gas kinetic energy have been discussed. Particular emphasis has been placed on absorption by the vibration rotation bands of diatomic molecules at high pressures. This high pressure absorption appears to offer efficient conversion of laser energy to gas translational energy. Bleaching and chemical effects are minimized and the variation of the total absorption coefficient with temperature is minimal. This latter feature is particularly advantageous from a systems design viewpoint.

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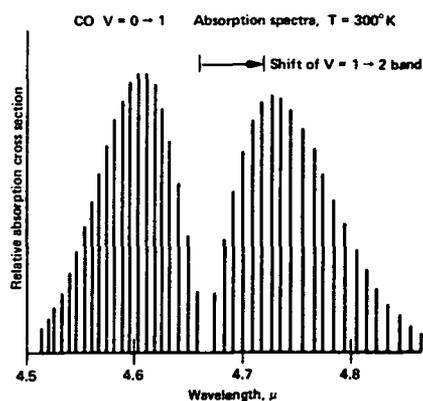


Figure 1.— Room temperature, low pressure absorption spectra of the  $V = 0 \rightarrow 1$  band of CO.

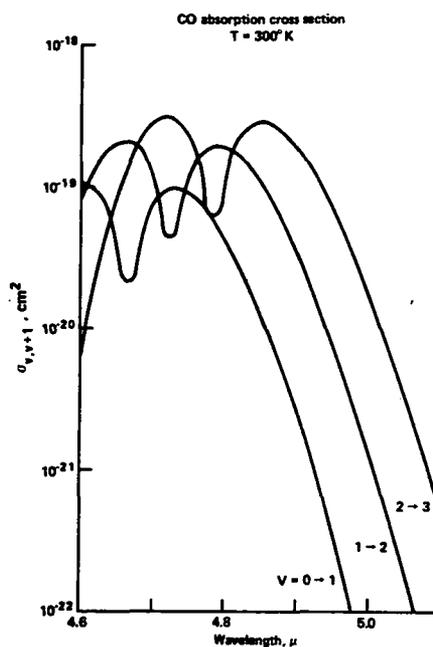


Figure 2.— Room temperature, high pressure absorption spectra of the  $V = 0 \rightarrow 1$ ,  $V = 1 \rightarrow 2$ , and  $V = 2 \rightarrow 3$  bands of CO.

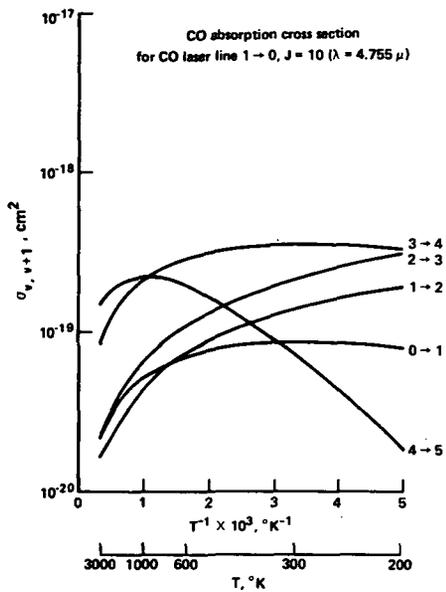


Figure 3.— Absorption cross section vs temperatures for the five lowest vibrational transitions in CO at a wavelength of 4.755  $\mu$ m.

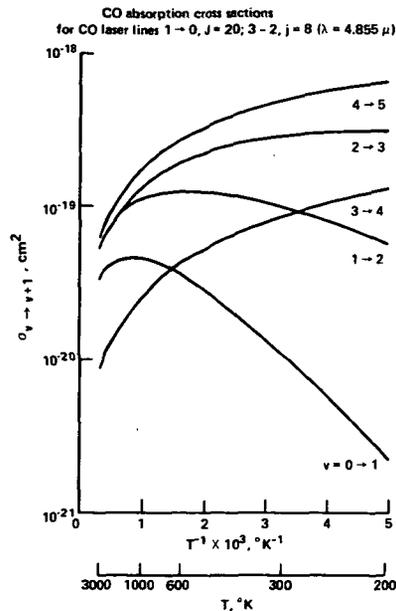


Figure 4.— Absorption cross section vs temperatures for the five lowest vibrational transitions in CO at a wavelength of 4.855  $\mu$ m.

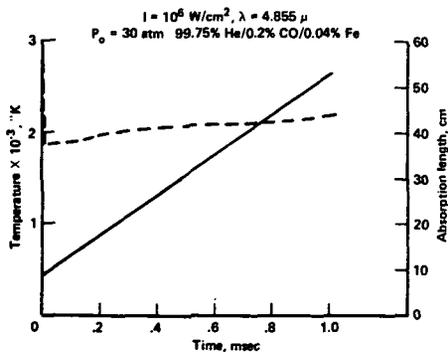


Figure 5.— Gas temperature and absorption length vs time; conditions as shown.

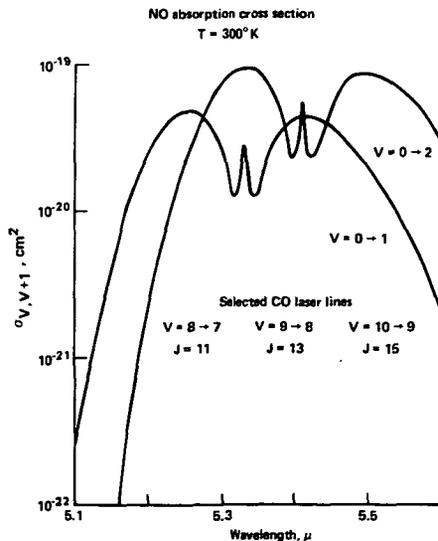


Figure 6.— Room temperature high pressure absorption spectra of the  $V = 0 \rightarrow 1$ ,  $V = 1 \rightarrow 2$  bands of NO.

## DISCUSSION

**Bob McKenzie, NASA Ames Research Center** – Are the dissociation energies of the hydrogen halides too low for you to consider?

**Answer:** There are two problems there. One is that all these hydrogen bearing species have relatively large rotational spacings, so one has to go to pressures like 100 atm to get this line overlap. However, I won't rule them out. Secondly, you're right, the dissociation energies are lower.

**Ernest Brock, Los Alamos Scientific Laboratory** – I don't want to be too nit-picking, but perhaps I will be! Even in the very low pressure-low temperature CO, there are quite a large number of lines in the V – V manifold that overlap to 0.1 to 0.5  $\text{cm}^{-1}$ .

**Answer:** Yes, as Lacina's recent publication indicates. Most of these are on higher vibrational levels, however. But your point's well taken – these accidental, rather than forced, overlaps do occur although the line centers don't match and hence the absorption can vary considerably.

**Ernest Brock, Los Alamos Scientific Laboratory** – The other comment I have, is that as one considers the uptake of energy in this system, one must keep in mind the relatively high cross sections for J – J transitions. In CO, for example, this is at 100 GHz and one should be concerned about self-trapping of the 100 GHz radiation.

**Answer:** I have assumed they are instantaneous in these calculations. One would have to do detailed kinetics to take it into account.

**Abe Hertzberg, University of Washington** – I would also note that the use of a Lorentzian is in error since there are indications that, if anything, it's super-Lorentzian.

**Answer:** Yes, that is correct.

**Mark Wrighton, M. I. T.** – What is the radiative probability from the upper excited state? That is, how long do they live?

**Answer:** The lifetime of the ground state, 1 – 0 transition, is like 33 msec, that might get down to 5 or 6 msec at high vibrational levels before it flattens out because of the energy dependence of the Einstein coefficient.

**Mark Wrighton, M. I. T.** – Is there any probability at all for a molecule to go from the fourth vibrational level down to the zeroth level?

**Answer:** Oh, that's very improbable. There is almost an order of magnitude between the fundamental and the first overtone. I might point out, however, about one-quantum transitions: anything radiated will be reabsorbed by the gas because all levels can absorb the wavelength.

**Malcolm Gower, NASA Ames Research Center** – Wouldn't you gain quite a bit by using a homonuclear molecule, like hydrogen and excite by stimulated Raman?

**Answer:** I would have thought the cross section was considerably lower.

**Malcolm Gower, NASA Ames Research Center** – Yes, except it becomes appreciable for high intensity laser sources.

**Abe Hertzberg, University of Washington** – Since inverse bremsstrahlung absorption works, and has a lot of nice properties, why not use it, in argon, say?

**Answer:** My point is that inverse bremsstrahlung under equilibrium conditions is very nonuniform in the first several thousand degrees of heating – that is, very temperature dependent.

**Abe Hertzberg, University of Washington** – Oh, I agree, but if I do have enough electrons, I could care less, I will absorb all the energy.

**Answer:** Well, it depends upon what the length of your system is. Of course at room temperature the absorption is zero. So, I'm saying that instead of using techniques which produce non-equilibrium electron concentrations, use this technique to get you to high enough temperatures so you can thermally ionize.

**Abe Hertzberg, University of Washington** – Well, being something of a practical engineer, I would suggest you use something like a flame thrower – the laser acts as the flame thrower.