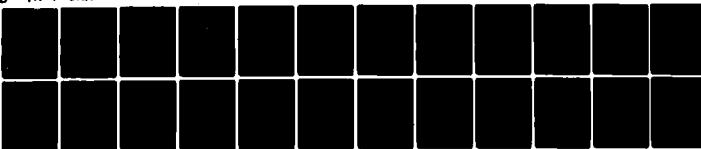


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there is no contribution to the angular momentum of CN other than the original angular momentum of the parent.

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ENERGY PARTITIONING IN THE DISSOCIATION OF CYANOGEN AT 193 NM

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ABSTRACT

The quantum state distributions of CN radicals produced in the photodissociation of C_2N_2 at 193nm have been measured. It has been found that the rotational distribution is the same for radicals formed in the $v''=0$ and $v''=1$ level. This has been interpreted to mean that the rotational motion is decoupled during the dissociation, from the vibrational and translational motion of the fragments. The partitioning of the $6795cm^{-1}$ of available energy between vibrational and translational motion in turn has been found to be consistent with a simple statistical model. The observed rotational distribution was reproduced by assuming that there is no contribution to the angular momentum of CN other than the original angular momentum of the parent.

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INTRODUCTION

There is a weak, highly structured absorption of cyanogen at 193 nm, that has been identified (1) as the $X^1\Sigma^+$ to $B^1\Delta_u$ transition. Since the $B^1\Delta_u$ state does not correlate with two CN radicals in the $X^2\Sigma$ state, dissociation can only occur via predissociation to the vibrational continuum of the ground state. This same continuum is responsible for the unimolecular thermal dissociation of C_2N_2 and its multiphoton infra-red dissociation. Thus dynamical studies of the partitioning of excess available energy among the two CN radicals can supply fundamental information about both of these processes.

The absorption coefficient to the $B^1\Delta_u$ state, about 10^{-19} cm^2 , is two orders of magnitude smaller than the one to the $C^1\Pi_u$ state, that occurs between 164 and 154 nm (2). This has effectively precluded both classical photochemical studies and modern dynamical studies of the single photon photodissociation of the $B^1\Delta_u$ state. The ArF laser has a sufficient flux, of about 10^{16} to 10^{17} photons/ cm^2 , to fragment a few percent of the cyanogen molecules, and thus makes it possible to carry out such dynamical studies despite the weak absorption coefficient at 193 nm.

Woo and Badger's analysis of the bands at 193 nm has shown that the absorption is at least the fourth member of a progression whose members are separated by about 2000 cm^{-1} (3). The frequency of this progression implies that it belongs to either the ν_1 or ν_3 mode of the excited state, which would primarily involve excitation of the CN bonds (4). In so far as a local mode picture is applicable, the

dissociation process must involve some excitation of the ν_2 mode, and thus flow of energy into the C-C bond. Thus the photolysis of cyanogen at 193 nm would involve predissociation, a conclusion which is also supported by the sharp structure of the cyanogen absorption spectrum in this region. Therefore one would expect that the amount of vibrational excitation in the product could be predicted from a statistical theory.

EXPERIMENTAL

The experimental apparatus consisted of a crude molecular beam source which was formed by a 1 mm i.d. hypodermic needle, spraying gas into a stainless steel chamber, pumped by a liquid nitrogen trapped 6" oil diffusion pump. The molecular beam flow was stabilized by a Granville-Phillips automatic pressure controller which maintained constant pressures in the scattering chamber. Experiments were done over a pressure range extending from 5×10^{-6} to 2×10^{-4} torr. The excimer laser beam entered the chamber from one side of the cell while the dye laser entered the cell from the other side. Both lasers were adjusted to be collinear, and crossed the beam spray at right angles. The smaller dye laser beam was completely imbedded within the excimer laser beam.

Fluorescent light, excited by the dye laser was imaged onto a filtered, bialkali photomultiplier tube, placed perpendicular to the plane defined by the laser and molecular beams. The lasers were pulsed at a rate of 10 Hz, with the excimer output coming 200 ns before the dye laser. This eliminated the effects of scattered excimer laser excited fluorescence from the cell windows, as well as precluding multiphoton up-pumping schemes based upon absorption of some combination of dye laser and excimer laser photons.

We observed no change in the nascent quantum state distributions of fragments, when the delay between lasers was varied, from zero, up to a few microseconds. An order of magnitude variation in both the background pressure of cyanogen, and the laser flux, also left the distribution of excess energy among

the fragments unchanged, although affecting the absolute magnitude of the signal in expected ways.

An attempt was made to detect any vibrationally cold CN ($A^2\Pi_1$) radicals produced by multiphoton absorption of ArF laser light. This was done by scanning the dye laser through the CN ($A^2\Pi_1 \rightarrow B^2\Sigma^+$) transition, a method which has been shown to be reasonably sensitive to small A state populations (5). Failure to detect any product in this first electronically excited allowed channel, sets an upper limit of less than 1% for excited product produced by multiphoton excitation.

Previous work showed that two photon sequential production of ($B^2\Sigma^+$) CN fragments from cyanogen depended on the square of the laser intensity at 193 nm (6). In this experiment, the laser intensities are comparable to those used in the previous work. The LIF signal measured by exciting the CN ($X^2\Sigma^+$) fragments when there is no delay between both lasers is at least ten times larger than the sum of the emission from directly produced ($B^2\Sigma^+$) radicals and scattered light from the excimer laser. Considering the larger volume illuminated by the eximer laser this sets an upper limit of 0.05% for the amount of $B^2\Sigma^+$ relative to $X^2\Sigma^+$ product.

Insertion of a beam flag resulted in a twofold decrease in signal intensity, but no change in the quantum state distribution of fragments. The very low pressures, at which the spectra were measured (2×10^{-5} torr), insure that there were no collisional effects. The fact that the same fragment distribution was measured for photolysis of the background, and of the beam with the background, may be taken to show that there is no appreciable cooling in the beam.

OBSERVATIONS AND DISCUSSION

Figure 1 is an example of the $\text{CN}(X^2\Sigma^+)$ excitation spectra obtained following photolysis of cyanogen at 193 nm. Here the delay between photolysis and probing is 200 ns, and the background pressure in the cell is 2×10^{-5} torr of cyanogen. One may parameterize the distribution of the $v''=0$ and $v''=1$ photoproduct by plotting the logarithm of the signal intensity divided by the Hoenl-London factor against the rotational energy of the fragment. The summed intensities of the P and R branches for eight excitation spectra are displayed in this manner in Figure 2.

Both the $v''=0$ and $v''=1$ distributions are well described by the same Boltzman distribution for a rigid rotor, with a parameterized "temperature", of about 900 K. The ratio of radicals produced in the first vibrational level, $v''=1$, to that produced in the $v''=0$ level is 0.35, as determined from the ratio of the intercepts in Figure 2, and after correction for the Franck-Condon factors.

As was mentioned above, at 193 nm cyanogen is excited to the $B^1\Delta_u$ state. It is clear that two CN radicals at infinite separation in the 2Σ state can not correlate with the $B^1\Delta_u$ state, which thus can only predissociate via a crossing into the vibrational continuum of the ground state.

The dissociation energy of cyanogen is $45,005 \pm 403 \text{ cm}^{-1}$, so that 6795 cm^{-1} of excess energy must be partitioned among the fragments (7). Since the cyanogen absorption spectrum (2,3) shows sharp structure at 193 nm, it is likely that the molecule undergoes several vibrations before dissociation. During these vibrations it is possible that randomization occurs so it is reasonable to try to explain the observed distribution of vibrational energy among the CN fragments on the basis of information theory (8). Following Kinsey and Levine (8), one may calculate the prior vibrational distribution for product CN to be

$$(1) \quad p^0(V,W|E) = 4/15 A_T A_R (E - E_V - E_W)^{5/2}$$

based on the assumption that the molecule is a rigid rotor harmonic oscillator, and where A_T is the translational state degeneracy, A_R allows for the sum over rotational states belonging to a particular vibrational level and V and W refer to the vibrational levels of the two diatomic fragments. E is the total amount of energy available. Figure 3 shows that the calculated prior distribution for $E = 6800 \text{ cm}^{-1}$ compares favorably with the observed vibrational distribution. Thus this analysis suggests that all of the available energy is preferentially distributed into translational and/or vibrational energy of the two CN fragments.

Energetically, $\text{CN}(X^2 \Sigma^+)$ radicals may be produced in the following vibrational channels:

- (a) $\text{C}_2\text{N}_2 + h\nu \rightarrow 2\text{CN}(X^2 \Sigma^+ v''=0)$
- (b) $\rightarrow \text{CN}(X^2 \Sigma^+ v''=0) + \text{CN}(2\Sigma^+ v''=1)$
- (c) $\rightarrow 2\text{CN}(X^2 \Sigma^+ v''=1)$
- (d) $\rightarrow \text{CN}(X^2 \Sigma^+ v''=2) + \text{CN}(X^2 \Sigma^+ v''=0)$
- (e) $\rightarrow \text{CN}(X^2 \Sigma^+ v''=2) + \text{CN}(X^2 \Sigma^+ v''=1)$
- (f) $\rightarrow \text{CN}(X^2 \Sigma^+ v''=3) + \text{CN}(X^2 \Sigma^+ v''=0)$

Since the amount of material in $v''=2$ and $v''=3$, as measured on the LIF spectra is below the limit of detection, if present at all, processes (d), (e) and (f) are not important. The problem is to decide between processes (b) and/or (c) as the source of $\text{CN}(2\Sigma^+ v''=1)$.

Remembering that the initial excitation involved at least four quanta of the ν_1 and/or ν_3 modes, where the energy is primarily localized in the CN bonds of cyanogen, and that the CN fragment LIF spectra showed no trace of $v''=2$ CN fragments, it is clear that there

has been redistribution of vibrational energy. The success of the prior distributions in reproducing the experimental results is a confirmation that some of the randomization of vibrational energy has occurred. Of course this reasoning is crude, and at best only qualitative, but it does provide a reasonable ex post facto explanation of the results.

On the basis of the success of the prior vibrational distribution in matching the data and the above line of reasoning one may suspect that process (c), which requires about the same amount of excess energy as process (d) is not important and that only processes (a) and (b) are significant photodissociation channels at 193nm. With this assumption and using the observed ratio of $[CN(v''=1)]/[CN(v''=0)] = 0.35$ one can compute the relative importance of reactions (a) and (b). The concentration of CN in the $[CN(v''=0)]$ state is given by $[CN(v''=0)] = N_a(v''=0) + N_b(v''=0)$, where N_a refers to the number of ground vibrational state radicals produced in process (a) and N_b to those produced in process (b). Since N_b must equal $[CN(v''=1)]$ simple arithmetic shows that the ratio of N_a/N_b is 1.86 and the quantum yield for process (a) is 0.52 and for process (b) it is 0.48. Thus within experimental error both are equally likely.

The fragment's nascent quantum state rotational distributions are identical for CN radicals created in the $v''=0$ and the $v''=1$ vibrational levels, so it is clear that in the dissociation process

the vibrational and rotational modes of motion are uncoupled. Moreover the success of the simple information theory approach implies that very little if any energy flows into rotation. In the absence of coupling between rotation and vibration conservation of angular momentum will "preserve" the rotational motion of the parent cyanogen molecule in its daughter CN fragments. One may calculate the probability of the linear cyanogen molecule being in any particular rotational state before absorbing a photon, and assign this probability to the two fragments each of which will have a rotational quantum number half of that of the parent molecule. The result of this procedure is displayed in Figure 4 as circles, while the data is shown as crosses. The excellent agreement with experiment confirms the model. To a good approximation such a procedure for cyanogen yields a Boltzmann distribution for the fragments with an apparent "temperature" of

$$(2) \quad T_{CN} = T_{C_2N_2} (B_{CN}/4B_{C_2N_2})$$

where T_{CN} is the parameterized temperature characterizing the CN fragment distribution, $T_{C_2N_2}$ is the temperature of the parent cyanogen, in this case 300K, and B is the appropriate rotational constant. Using the values of $B_{CN} = 1.89 \text{ cm}^{-1}$ (9) and $B_{C_2N_2} = 0.157 \text{ cm}^{-1}$ (10) one would predict that $T_{CN} = 900 \text{ K}$, which is exactly what is found.

This also explains why the surprisals calculated in Ref. 3 for the product radicals produced in the $C^1\Pi_u$ state had a positive slope. There it was assumed that all of the excess energy was available to

rotation. However, in that case there may be coupling between vibration and rotation, as shown by the difference between the measured rotational temperatures for CN radicals produced in different vibrational levels. Clearly not all of the excess energy is available to be distributed into rotation and there is a significant contribution to the rotational excitation of the fragments from the thermal excitation of the parent cyanogen.

CONCLUSIONS

Laser excitation of the $B^1\Delta_u$ state of cyanogen, followed by LIF detection of the quantum state distribution of the fragments has shown that this state predissociates into two ground state CN radicals. This predissociation must occur via the vibrational continuum associated with the C-C motion of the ground state of cyanogen. It has been shown that the quantum yield for simultaneous production of two ground state CN radicals is one half, and that the quantum yield for the production of one CN radical in the $v''=0$ level and the other in the $v''=1$ level is also one half.

The rotational energy of the fragments has been demonstrated to be a remnant of the rotational motion of the parent cyanogen. This implies that the geometry of the $B^1\Delta_u$ state is linear, in agreement with the previous spectroscopic assignment.

Good agreement has been obtained when the observed vibrational population was compared to a population based upon randomization of the available energy in either the vibrational or translational motion of the fragments. The rotational motion is however, essentially decoupled from the other two so that angular momentum is conserved by folding the original angular momentum of the C_2N_2 molecule into the angular momentum of the fragments.

The techniques that have been described are useful for illumi-

nating the processes that occur during photodissociation. The C_2N_2 molecules allow one to look not only at photodissociation dynamics but also the dynamics of multiphoton i.r. photolysis as well as unimolecular reactions. In the former case we are preparing the superexcited molecule at a fixed and known energy above the ground state continuum, while in i.r. multiphoton processes the molecule is excited by driving it through this quasi-continuum state. In both cases unimolecular decay of this super excited molecule then occurs. One would therefore, predict that i.r. multiphoton dissociation of C_2N_2 should lead to the same type of dynamics as the photodissociation of the $B^1\Delta_u$ of C_2N_2 .

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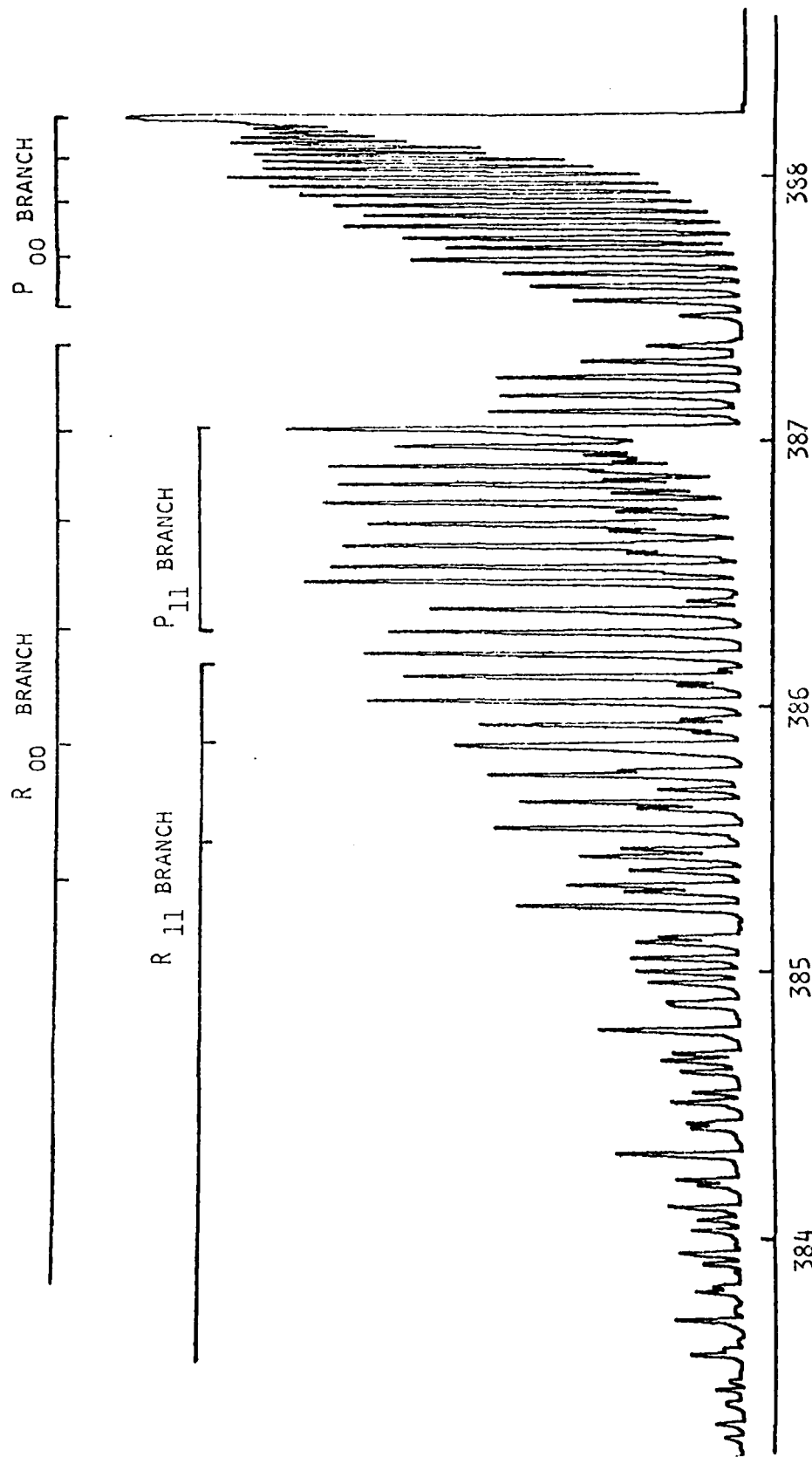
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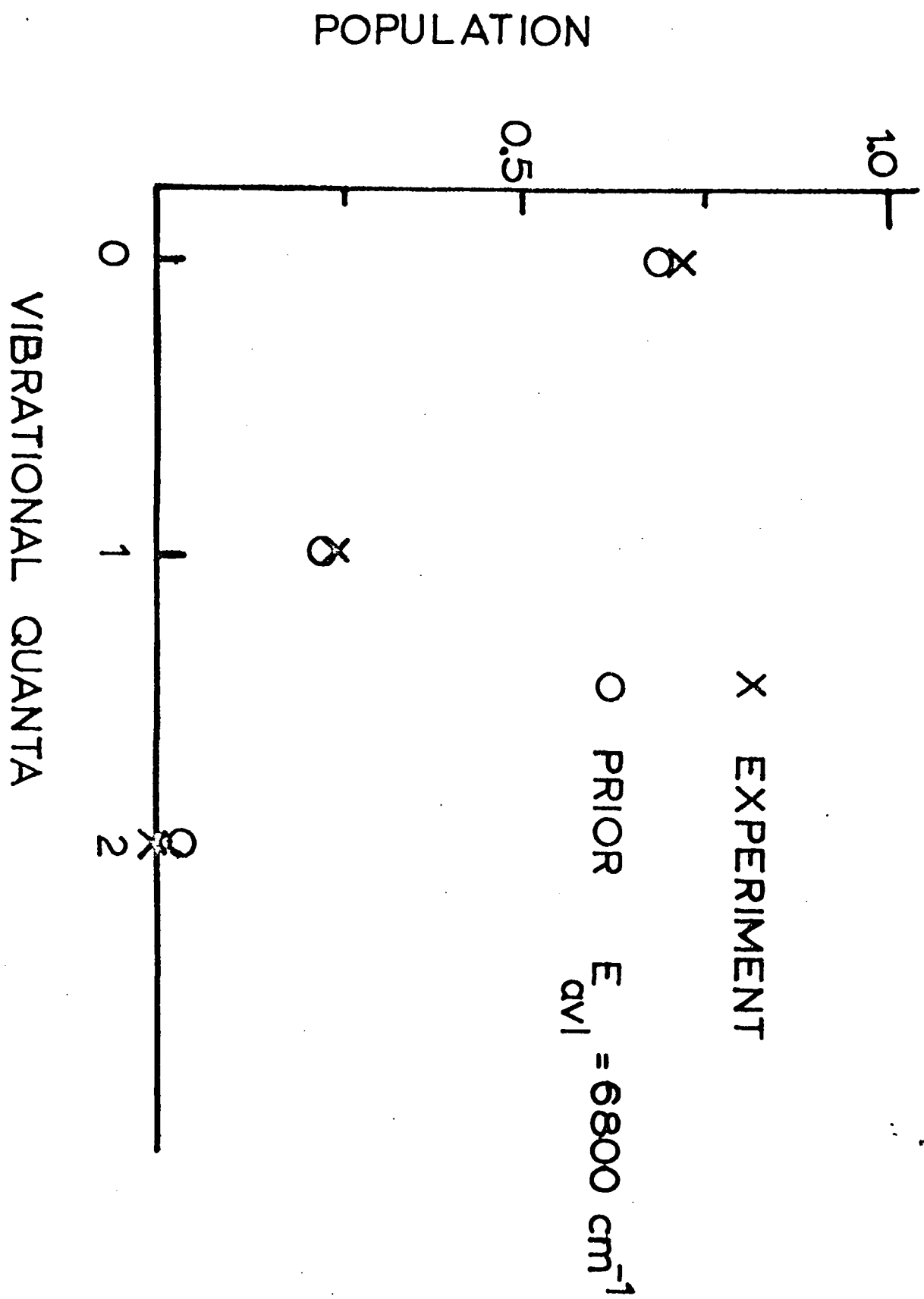
- Figure 1: LIF excitation spectrum of the $CN(X^2\Sigma^+)$ fragments from photolysis of cyanogen at 193nm. The background pressure of cyanogen in the cell was 2×10^{-4} torr and the delay between the excimer laser and dye laser was 200ns.
- Figure 2: Averaged values of $\ln [I(N'')/(N'+N''+1)]$ plotted against the rotational energy of CN fragments with rotational quantum number N'' . $I(N'')$ is the measured LIF signal from the state N'' and N' labels the emitting state.
- Figure 3: Vibrational prior distributions and experimental results plotted against the number of vibrational quanta in the fragments. Crosses represent data, circles represent the calculated prior distribution with an available energy of 6800 cm^{-1} .
- Figure 4: Rotational distribution of fragments plotted against rotational energy. Crosses represent the data and circles are the results of a model described in the text.

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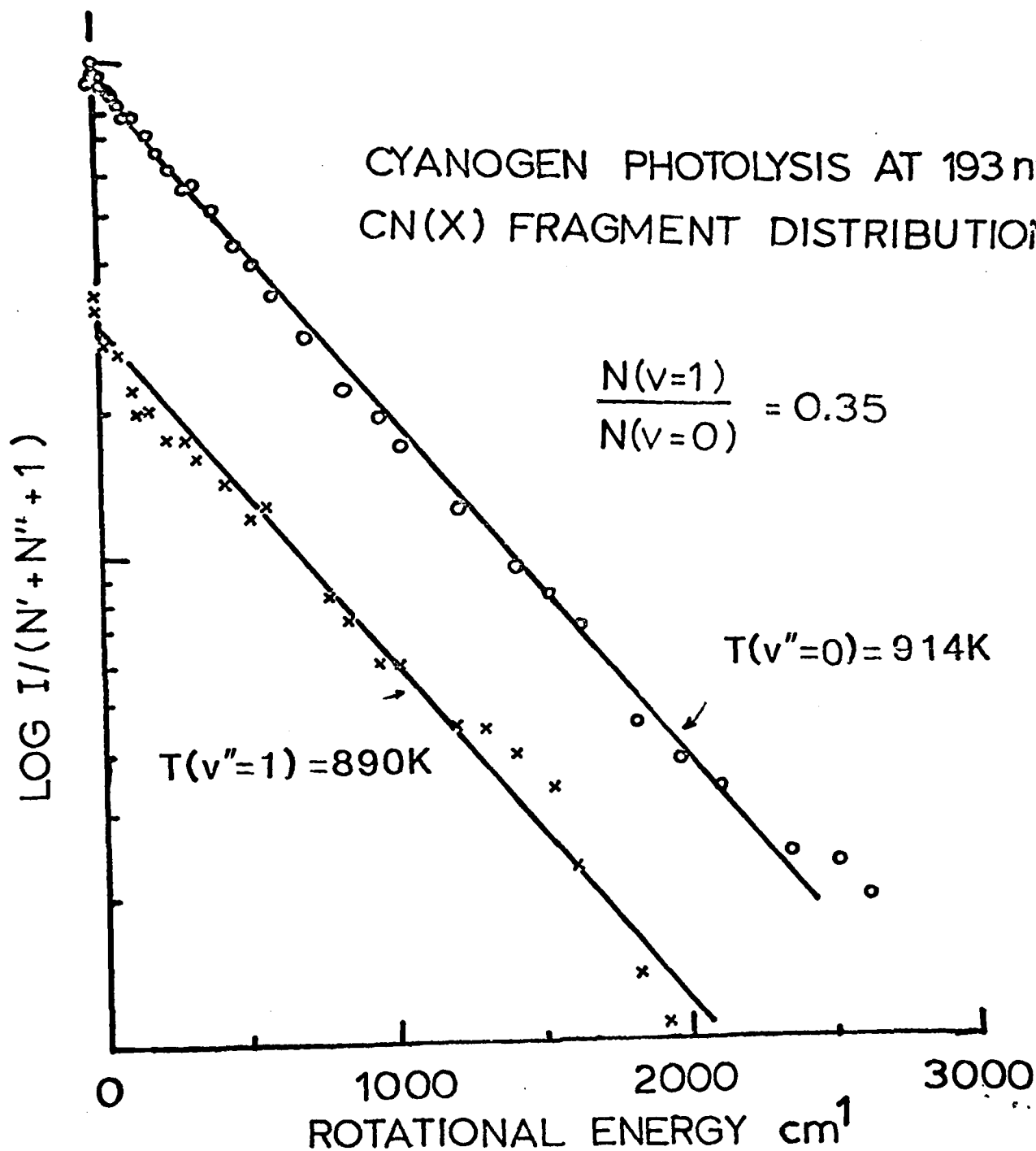
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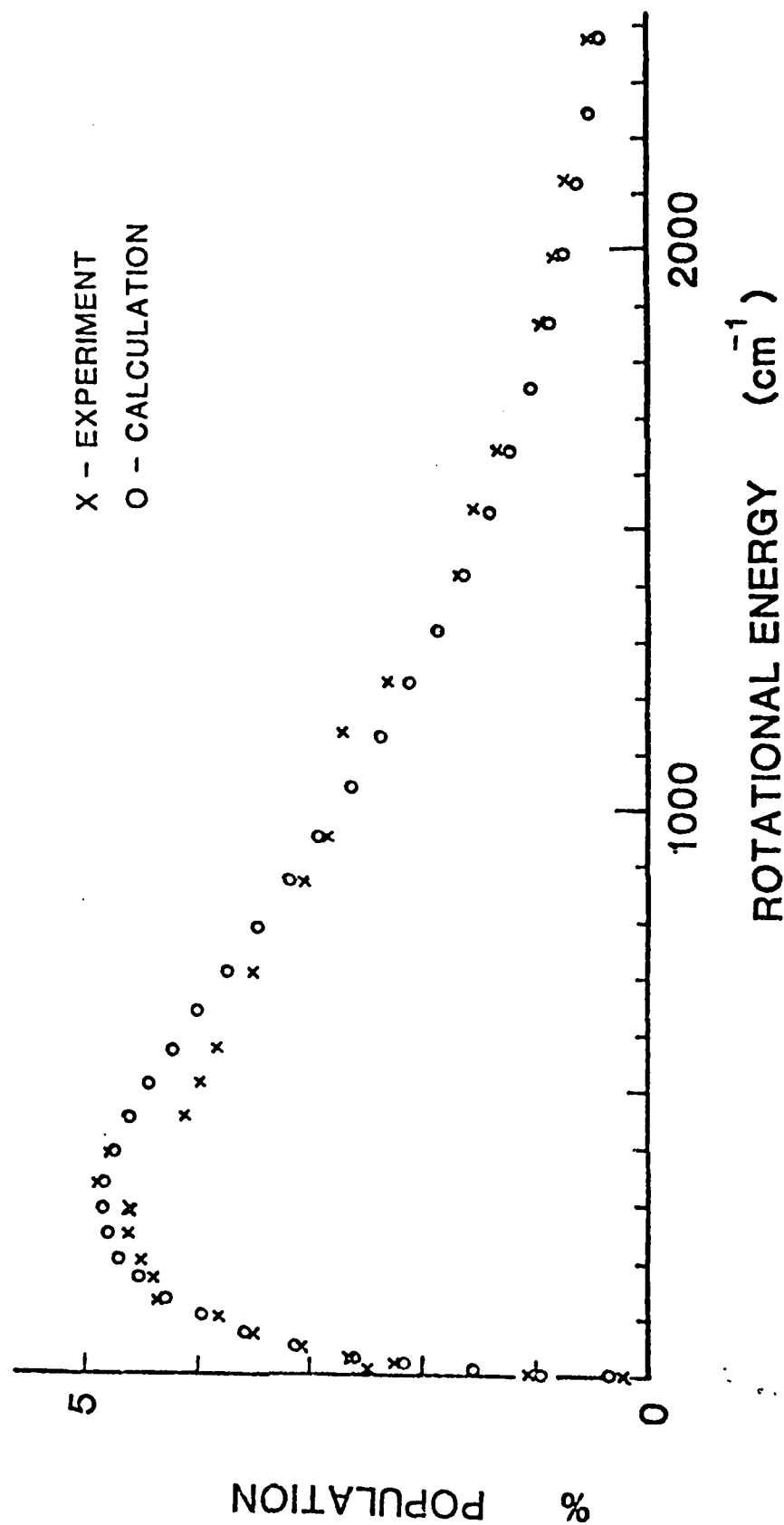




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