Dicke Narrowing and Pressure Broadening in the Infrared Fundamental Band of HCl Perturbed by Ar

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Dicke narrowing due to state preserving but velocity changing collision has been observed in several high-J vibration-rotation transitions of the fundamental band of HCl with Ar as the collision partner. Because of the long wavelength of radiation and the small diffusion coefficient, the narrowing occurs at a relatively low pressure of 200 Torr. A difference frequency laser system was employed for the studies of linewidths and lineshapes over the temperature range of 297–420 K. The results were analyzed using the diffusion model.

I. INTRODUCTION

The effect of collisions on the Doppler widths of spectral lines was originally discussed in 1953 by Dicke (1). He showed that a substantial narrowing of the linewidth is expected if (a) the collisions do not affect the quantum state of the absorbing or radiating system and (b) the mean free path is smaller than the wavelength of the interacting radiation. This effect was first experimentally demonstrated for the hyperfine transition in the ground state of atomic hydrogen by Wittke and Dicke (2). Since then many papers have been published on the theoretical and experimental aspects of this interesting effect. The original intuitive picture by Dicke has been more formally analyzed by Galatry (3) who considered soft collisions and by Nelkin and Ghatak (4) and by Gersten and Foley (5) who considered hard collisions. Rautian and Sobel’man (6) and Murray and Javan (7) considered both soft- and hard-collision models and compared theoretical and experimental results. The experimental demonstration was initially performed in the radiofrequency or microwave region for hyperfine transitions of atoms (H, Na, Cs, Rb; 2, 8–11). The two required conditions (a) and (b) mentioned earlier are more easily satisfied for these cases. Later the experiment was extended to optical regions and to molecules; in these cases it is more difficult to satisfy the two conditions because of the fast rotational relaxation and the short wavelength of radiation. Molecules with widely separated rotational levels and high pressure were used to match the required conditions. Dicke narrowing was observed in both spontaneous and stimulated Raman spectra, Rayleigh scattering as well as CARS for the vibration-rotation transitions of H₂, HD, and D₂ (12–20). It was also observed in the infrared

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spectra of quadrupole-induced vibration–rotation transitions of \( \text{H}_2 \) and \( \text{D}_2 \) (21–23) and of rotational transitions of \( \text{H}_2 \) and \( \text{H}_2\text{O} \) (24, 25). More recently, Pine has carried out a detailed study on the collisional narrowing of the high-J vibration–rotation lines of HF (26). Even for low rotational levels of polar molecules such as \( \text{NH}_3 \) (27) and \( \text{CH}_3\text{CN} \) (28) in which large pressure broadening masks the collisional narrowing, the effect of the latter is revealed after careful analysis of the linewidth.

In the present paper we report our study of the phenomenon of collisional narrowing in the infrared vibration–rotation spectrum of HCl in Ar (29). The spectrum appears at 3.5 \( \mu \text{m} \) with the band origin at 2886 cm\(^{-1} \) (30). As in the case of Pine’s work on HF, the use of infrared spectrum affords a good possibility for a detailed study of Dicke narrowing because the condition (b) that the mean free path is less than the wavelength is fulfilled at a relatively low pressure of tens of torr [sic]Cells, contrary to the optical case for which much higher pressure is needed.

II. EXPERIMENTAL DETAILS

The difference frequency laser system developed by Pine (31) generates frequency tunable infrared radiation between 2400 and 4400 cm\(^{-1} \) and provides a useful source for studies of Doppler-limited absorption spectrum. Such a source constructed in our laboratory (23, 32, 33) has been employed to study the line widths of several fundamental vibration–rotation transitions of HCl with Ar atoms as collision partners. Other rare gas atoms such as Ne and Xe were also used as collision partners but did not produce the clear narrowing as in the case of Ar. This is due to the fact that a collision partner with equal mass is most effective in changing the velocity through collisions. Thus, the combinations of HF–Ne used by Pine (31) and HCl–Ar used in this work give the most prominent narrowing. We used the \( R(9), R(11), R(13), R(15), R(17) \) lines for the studies of lineshape. The \( R(9) \) and \( R(11) \) lines were studied at room temperature and the \( R(15) \) and \( R(17) \) lines were studied at elevated temperatures (370 and 420 K, respectively). The \( R(13) \) line was studied at both room temperature and at 420 K to study the temperature dependence. For studying the \( R(9) \) transition which is very strong and requires little sample of HCl, the less abundant isotropic species H\(^3\)Cl was used for ease of accurate pressure adjustment. The condition (a) required for narrowing, that the HCl molecules conserve their rotational quantum state while changing velocities due to collisions, is more easily satisfied for higher-J transition; the rotational energy spacing for the \( R(9) \) line is \( \sim 200 \text{ cm}^{-1} \) while it is \( \sim 370 \text{ cm}^{-1} \) for the \( R(17) \) line much larger than \( kT \). Thus higher-J transitions give much larger narrowing than low-J transitions as shown later (for example, in Fig. 4).

The experiments were carried out using absorption cells with lengths of 50 cm and 2 m, and the pressure of HCl was varied from a fraction of 1 Torr to the maximum of 12 Torr, depending on the transition. The condition was chosen such that the absorption was \( \sim 20–30\% \) due to pure HCl, that is, prior to the addition of Ar. Argon pressure was varied from 20 Torr to one atmosphere and the pressure was read on a Baratron gauge. Infrared spectral lines of \( \text{H}_2\text{O} \) were used for calibration. The linewidth measurements were made with the help of 300-MHz interference markers generated from a Toepel spectrum analyzer which monitors the dye laser radiation. The accuracy of the linewidth measurement was about ±5 MHz for not too broadened lines. The
inaccuracy of drawing the base line sometimes gave large uncertainty in the observed linewidth, especially in the case of the Lorentzian lineshape which exhibits long tails. This occurs at high pressures of Ar. The error caused by this is \( \sim 2\% \) for the high pressure case and less for lower pressure.

III. OBSERVED RESULTS

Figure 1 shows a typical spectral narrowing and the variation of lineshape of the \( R(13) \) HCl line due to the addition of Ar gas. The narrowing of the spectral line and the transition of the lineshape from the usual Doppler-broadening Gaussian shape to the Dicke-narrowed Lorentzian shape is clearly seen.

Table I summarizes the spectral lines of HCl used for the study, experimental conditions, and observed and calculated linewidths for pure HCl gas. The table provides the basic data for our measurement and gives an idea of the accuracy of our measurements. The calculated linewidths were obtained using the formula

\[
\Delta \nu = \left( \Delta \nu_D + \frac{\Delta \nu_p}{\sqrt{\eta}} \right),
\]

where \( \Delta \nu_D \) is the Doppler half-width and \( \Delta \nu_p \) is the pressure broadening. This formula is obtained from the usual Voight profile

\[
I(x) = \frac{1}{\sqrt{\pi \eta}} \int_{-\infty}^{\infty} \frac{\exp[-(x-y)^2/\eta^2]}{1+y^2} dy,
\]

with \( \eta = \Delta \nu_D / \Delta \nu_p \) by taking the limit \( \eta \gg 1 \).

The \( R(13) \) line of HCl was the most extensively studied transition and the results are shown in Fig. 2. It is seen that a reduction in half-width at half-height of about

![Graph showing typical lineshapes for the HCl R(13) line of the fundamental band. HCl gas with 1 Torr pressure was used with (upper trace) and without Ar gas.](image-url)
DICKE NARROWING IN HCl

TABLE I

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Line</th>
<th>Temperature (K)</th>
<th>Pressure (Torr)</th>
<th>δν&lt;sub&gt;obs&lt;/sub&gt; (MHz)</th>
<th>δν&lt;sub&gt;calc&lt;/sub&gt; (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H&lt;sup&gt;37&lt;/sup&gt;C&lt;sub&gt;1&lt;/sub&gt;</td>
<td>R(9)</td>
<td>297</td>
<td>0.3</td>
<td>93.6</td>
<td>93.2</td>
</tr>
<tr>
<td>H&lt;sup&gt;35&lt;/sup&gt;C&lt;sub&gt;1&lt;/sub&gt;</td>
<td>R(11)</td>
<td>297</td>
<td>0.5</td>
<td>96.5</td>
<td>95.5</td>
</tr>
<tr>
<td>H&lt;sup&gt;36&lt;/sup&gt;C&lt;sub&gt;1&lt;/sub&gt;</td>
<td>R(13)</td>
<td>297</td>
<td>3</td>
<td>97.5</td>
<td>98.3</td>
</tr>
<tr>
<td>H&lt;sup&gt;36&lt;/sup&gt;C&lt;sub&gt;1&lt;/sub&gt;</td>
<td>R(13)</td>
<td>370</td>
<td>1.8</td>
<td>112</td>
<td>112</td>
</tr>
</tbody>
</table>

*The observed widths are derived from an average of five measurements which had a scatter of <±5 MHz.

25% is seen for the Ar pressure of ~200 Torr. The analysis of the observed widths for this line as well as for other lines followed the standard procedure in which an assumed diffusion coefficient and the calculated Dicke-narrowed Doppler widths are varied until the pressure-broadening width, i.e., the difference between observed widths and Dicke-narrowed Doppler widths, follows a straight line as a function of Ar pressure. When the small effect of the pure HCl pressure broadening is subtracted, the straight line passes through the origin as in Fig. 2. Since the diffusion model fails at low pressures, the derived pressure-broadened widths deviate from the straight line. In our experiment, this deviation was seen for pressures less than 150 Torr. The data in this area were not used in the analysis. An alternative way to look at the variation in

![Graph](image-url)

**Fig. 2.** Observed linewidths for the R(13) line as a function of Ar pressure and their decomposition to Dicke narrowing and pressure broadening using the diffusion model Eq. (1).
linewidths is to plot the peak height of absorption as a function of Ar gas pressure as in Fig. 3. The peak height initially increases with the pressure because of collisional narrowing, passes a maximum, and then decreases because of pressure broadening.

Different transition lines give different linewidth dependencies on pressure because of different pressure broadening parameters. The higher-$J$ lines give lower pressure broadening due to more separated rotational energy levels and thus show more marked Dicke narrowing. This situation was clearly seen in our observations, which are plotted in Fig. 4. The variation of widths is also dependent on temperature. The observed dependence is shown in Fig. 5 in which measured widths of the $R(13)$ lines are plotted for three temperatures 297, 370, and 420 K. It was noted that at the high-pressure region the widths at 420 K fall between those at 297 and 370 K. The higher temperature
increases the Doppler width but decreases the pressure broadening because of the $T^{-0.7}$ dependence of the pressure broadening on temperature (35) for the $1/r^6$ interaction between HCl and Ar. Thus, at higher temperature the linewidths start high at low pressure but decrease more rapidly, and this explains the data given in Fig. 5.

The observed variation of lineshape for different pressures is shown in Fig. 6. The dots in the figure represent calculated Lorentzian curve fits to the lines. It is seen that the lineshape changes from a Gaussian shape for pure HCl to a Lorentzian shape at high pressure; above the pressure of $\sim 250$ Torr, the shape was completely Lorentzian within the experimental uncertainties. This occurs when the ratio of pressure and diffusion coefficient $(P/D)$ reaches a certain value (7). This is reached at a much lower pressure for the HCl:Ar mixture than for HF:Ne because the diffusion coefficient for the former is smaller than for the latter by a factor of $\sim 2.7$. Our results did not discriminate between hard- and soft-collision models and did not detect the small asymmetry reported by the more accurate measurements of Pine (26).

IV. ANALYSIS

The analysis of the pressure dependence of linewidths was carried out using high-pressure results assuming the linewidths at high pressure to be Lorentzian and using the diffusion model. The observed widths (half-width at half-maximum) were fitted to the expression

$$\Delta \nu = \frac{AT^\alpha}{P} + BPT^{-\beta},$$

(1)

where $P$ and $T$ are pressure and temperature and $A$, $\alpha$, $B$, and $\beta$ are parameters to be determined from the observed results. The first term, inversely proportional to pressure, represents the Dicke narrowing and the second term, pressure broadening. The simple addition of two terms is justified because both terms make the absorption Lorentzian. Note that we factor out the temperature dependence explicitly in the Dicke-narrowing coefficient and the pressure-broadening coefficient. The coefficient of $1/p$ and $p$ are related to the standard notation as
\[ AT^\alpha = 2\pi D/\lambda^2, \]  

(2)

where \( D \) is the diffusion coefficient and \( \lambda \) is the wavelength (2, 3) and

\[ BT^{-\beta} = \Delta \nu_p, \]  

(3)

where \( \Delta \nu_p \) is the pressure-broadening parameter (35). By fitting the pressure dependence of the observed linewidths in Eq. (1), we obtained the values of \( AT^\alpha \) and \( BT^{-\beta} \) as shown in Table II. Since the diffusion model does not apply to low-pressure cases, observed results with high pressure (\( p \geq 250 \) Torr) were used for the determination.

Equation (1) shows that the linewidth is at its minimum when the two terms on the right-hand side are equal, that is,

\[ p = \sqrt{\frac{A}{B}} T^{(\alpha+\beta)/2}. \]  

(4)

Figure 2 shows that the minimum indeed occurs when the curves for the two terms cross. The minimum width is

\[ \Delta \nu_{\text{min}} = 2\sqrt{AB} T^{-(\alpha-\beta)/2}. \]  

(5)

Since the temperature dependence of \( \Delta \nu_{\text{min}} \) is not much different from that of the Doppler width (\( \propto T^{1/2} \)), the ratio of \( \Delta \nu_D/\Delta \nu_{\text{min}} \) is almost independent of temperature.
TABLE II

Dicke-Narrowing Coefficient $AT^n$ (in MHz atm) and Pressure-Broadening Parameter $BT^n$ (in MHz/Torr) Determined from the Observed Linewidths

<table>
<thead>
<tr>
<th>Line</th>
<th>Temperature (K)</th>
<th>$AT^n$ (MHz atm,)$^a$</th>
<th>$BT^n$ (MHz/Torr)$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>R(9)</td>
<td>297</td>
<td>8.25 (11)</td>
<td>0.346</td>
</tr>
<tr>
<td>R(11)</td>
<td>297</td>
<td>8.25 (11)</td>
<td>0.250</td>
</tr>
<tr>
<td>R(13)</td>
<td>297</td>
<td>8.25 (11)</td>
<td>0.197 (0)</td>
</tr>
<tr>
<td>R(15)</td>
<td>370</td>
<td>12.5 (-4)</td>
<td>0.169 (-4)</td>
</tr>
<tr>
<td>R(15)</td>
<td>420</td>
<td>16.0 (-8)</td>
<td>0.162 (1)</td>
</tr>
<tr>
<td>R(15)</td>
<td>352</td>
<td>11.5 (-1)</td>
<td>0.154</td>
</tr>
<tr>
<td>R(17)</td>
<td>420</td>
<td>18.0 (12)</td>
<td>0.121</td>
</tr>
</tbody>
</table>

$A = 6.40 \times 10^{-5}$ MHz atm/deg$^a$ \hspace{1cm} $a = 2.07$

$B = 5.35$ MHz deg$^a$/torr \hspace{1cm} $b = 0.58$

*Numbers in parentheses indicate observed minus calculated values in the last digits using the parameters given in this table.

A. Dicke-Narrowing Coefficient

The observed Dicke-narrowing coefficients $AT^n$ listed in Table II were used to determine $A$ and $\alpha$ on the assumption that they are independent of $J$ (26). Since the Dicke-narrowing parameter is due to velocity changes of molecules accompanying the HCl:Ar collision, such as assumption is probably justified. The parameters obtained are listed in Table II. These values and Eq. (2) give the diffusion constant $D$ of HCl in Ar at room temperature to be $0.140 \text{ cm}^2 \text{ sec}^{-1}$ which is comparable to the self-diffusion coefficients of HCl and Ar of $0.1246$ (at $295 \text{ K}$) and $0.156$ (at $273 \text{ K}$), respectively (37).

The observed temperature dependence of $T^{2.07}$ for the Dicke-narrowing coefficient must arise from the temperature dependence of diffusion coefficient $D$. The latter is calculated theoretically (38) to be

$$D \propto T^{4+S},$$

where

$$S = \frac{1}{2} + \frac{2}{n-1}$$

for the intermolecular interaction potential of $1/r^n$ dependence. The dependence of the HCl:Ar interaction gives the temperature dependence of $T^{1.9}$ which is in reasonably good agreement with the observed $T^{2.07}$. 
The pressure-broadening part of the linewidths obtained from the analysis for the R(9), R(11), and R(13) transitions at room temperature and for the R(13) and R(17) transitions at 420 K as a function of Argon pressure is shown in Fig. 7. The pressure-broadening parameters (Δν)p determined from the slopes of these straight lines are plotted in Fig. 8 together with previous experimental results (39, 40) as a function of J. It is noted that Δνp decreases rapidly with increasing J values because of more widely spaced rotational levels and accordingly lower rotational transition probabilities. On the average an HCl molecule in the J = 18 level stays in the quantum state 20 times longer than an HCl molecule in the J = 1 level. Assuming that the contribution to linewidth comes mainly from state-changing collisions in which R ↔ T energy transfer is realized, the observed results were fit to the simplified formula

$$Δν_p = Δν_0 \exp(-αJ).$$  

(7)

Fig. 7. Pressure broadening versus Ar pressure for the R(9), R(11), and R(13) lines at 297 K and the R(13) and R(17) lines at 420 K.

Fig. 8. Pressure-broadening parameters for different R(J) transitions at room temperature.
A curve drawn with $\Delta \nu_0 = 2 \text{ MHz/Torr}$ and $\alpha = 0.190$ is shown in Fig. 7. The curve fit well to observed results between $J = 0$ and 11 though the experimental results are significantly higher than the "theoretical" curve values for $J \geq 13$. Whether this deviation is due to phase-changing collisions or reorientation remains to be seen. When the earlier observed data for HCl:Kr and HCl:Xe (39) are reanalyzed using Eq. (7), we obtain the $\alpha$ values of 0.13 and 0.10, respectively. Thus for a heavy-collision partner, the value of $\alpha$ is close to $2B/kT = 0.10$. It is also noted that the values of $\alpha$ are inversely proportional to the polarizabilities which are 16.3, 24.6, and 40 (in units of $10^{-25} \text{ cm}^3$) for Ar, Xe, and Kr, respectively.

Several theoretical papers on HCl:Ar collisions have been published. The calculated theoretical values are listed in Table III together with the experimental results. None of the theoretical treatments seem to explain satisfactorily the observed results in the full range of $J$.

### TABLE III

Experimental and Theoretical Pressure-Broadening Parameters $\Delta \nu_p$ for HCl in Ar

<table>
<thead>
<tr>
<th>R(J)</th>
<th>RERW</th>
<th>LPB</th>
<th>Present Work</th>
<th>Theoretical</th>
<th>BIL</th>
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</thead>
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<td></td>
<td></td>
<td></td>
<td></td>
<td>RGG TH</td>
<td>HT</td>
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<tr>
<td>0</td>
<td>2.53</td>
<td>2.05</td>
<td>1.62</td>
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<td>1</td>
<td>1.68</td>
<td>1.38</td>
<td>1.42</td>
<td>1.34</td>
<td>1.50</td>
</tr>
<tr>
<td>2</td>
<td>1.76</td>
<td>1.20</td>
<td>1.14</td>
<td>1.18</td>
<td>1.24</td>
</tr>
<tr>
<td>3</td>
<td>1.54</td>
<td>1.12</td>
<td>0.91</td>
<td>1.03</td>
<td>1.03</td>
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<tr>
<td>4</td>
<td>1.10</td>
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<tr>
<td>5</td>
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<tr>
<td>8</td>
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<tr>
<td>9</td>
<td>0.43</td>
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<td>10</td>
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<td>12</td>
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<td>13</td>
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<td>16</td>
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<tr>
<td>17</td>
<td></td>
<td>0.101</td>
<td></td>
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</tr>
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</table>

Note. RERW = Rank, Eastman, Rao, and Wiggins (39); LPB = Levy, Poillet-Mariel, and Boulet (40); RGG = Robert, Giraud, and Galatry (41); TH = Tipping and Herman (42); BIL = Boulet, Isnard, and Levy (43); HT = Herman-Tipping potential, B = Buckingham potential; LS = Leavit (44).
The temperature dependence of the pressure-broadening parameter was determined from the observed values for the R(13) line listed in Table II to be $T^{-0.58}$. This is in reasonable agreement with the $T^{-0.7}$ calculated from $(Δν)T ∝ T^{-(n+1)/2(n-1)}$ for the $1/r^n$ potential with $n = 6$ (35). The temperature dependence is smaller than those determined earlier for the HCl:Ar system by Van Aalst et al., $T^{-0.96}$ (calculated from the temperature dependence of the cross section given in Ref. (45)), by Petrov, $T^{-0.69}$ (determined from the graph given in Ref. (46)), and by Miziolek, $T^{-1.67}$ (calculated from Table IV of Ref. (47)). It is larger than that given by Houdean et al., $T^{-0.2}$ (48).

V. CONCLUSION

We have measured and analyzed the lineshapes of high-J vibration–rotation lines of HCl perturbed by Ar gas. Because of the long wavelength (3.5 μ) and small diffusion coefficient of the HCl:Ar system, we could accurately cover the entire region where the linewidths decrease because of Dicke narrowing and then increase because of pressure broadening. We used the simple diffusion model and the observed results for the high-pressure range (i.e., after the linewidth minimum is reached) to determine the Dicke-narrowing coefficients and the pressure-broadening parameter and their temperature dependence which is between the self-diffusion parameters 0.125 cm². The former gave the diffusion parameter of HCl in Ar of 0.140 cm² sec⁻¹ which is between the self-diffusion parameters 0.125 cm² sec⁻¹ of HCl and 0.156 cm² sec⁻¹ of Ar. The temperature dependence of the Dicke-narrowing parameter $T^{-2.07}$ is in agreement with the theoretical temperature dependence. The observed pressure broadening together with previous values fit reasonably well to the $exp[-αJ]$ curve although the observed values were higher for very-high-J values. The observed values in this paper do not agree with any theoretical calculations published earlier. The accuracy of our measurements for the HCl:Ar system is higher than the previous cases probably because of the use of monochromatic tunable radiation sources and the careful separation of the narrowing effect. Compared with the recent very accurate measurements by Pine of the HF–Ne system (26), our results did not discriminate the subtle difference between the hard- and soft-collision models and did not detect the asymmetry which gives more information on the Dicke-narrowing coefficients and pressure-broadening parameters.

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