

ROTATIONAL EXCITATION OF MOLECULAR IONS BY ELECTRON IMPACT UNDER INTERSTELLAR CONDITIONS

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ABSTRACT

A generalized expression for the rate coefficient for rotational excitation of molecular ions by electron impact under interstellar conditions has been obtained from the first order perturbation theory. The expression has been obtained by considering only the electron-dipole term of the interaction potential which is the most dominant term. The effect of short range and electron-quadrupole interactions has been assessed. The importance of electron-ion collisions in relation to H_2 -ion collisions in the interpretation of the spectral data obtained for ions from interstellar sources has also been assessed.

Subject headings: interstellar: molecules — molecular processes

1. INTRODUCTION

Early interest in the rotational excitation of molecular ions by electrons was closely linked to the fact that such processes are the most effective mechanism of the energy loss of low energy electrons in a partially ionized molecular gas. This explains the preoccupation in the past with the excitation of H_2^+ and its isotope HD^+ (Stabler 1963; Sampson 1965). Recently, however, molecular ions have been discovered in dark interstellar clouds, the most important of which are N_2H^+ and HCO^+ . A knowledge of their abundances and rotational excitation cross sections is important for a satisfactory interpretation of the spectroscopic data obtained from these ions in interstellar sources and also for the critical evaluation of models proposed for molecule formation in dark clouds.

The intensities of the observed rotational lines of these ions are determined by a competition between the collisional and radiative processes. Since H_2 is the most abundant species in dark clouds, collisions with H_2 are generally considered. However, the presence of small quantities of electrons in such clouds is now well established. Since e -ion rates are expected to be much higher than the corresponding H_2 -ion rates, the e -ion collisions may be important for a proper interpretation of the data. It may be mentioned that for neutral molecules the rates for H_2 -molecule and e -molecule collisions have been calculated (Green and Thaddeus 1976; Green and Chapman 1976; Dickinson and Richards 1975; Saha *et al.* 1980) and the relative effects of the two types of collisions on line intensities have been assessed (Dickinson *et al.* 1977, Bhattacharyya and Barua 1981a). For ions detected in dark clouds such theoretical studies have just been started, and considerable amount of work remains to be done.

Since microwave and radio emission lines from interstellar clouds can be observed only from molecules with permanent dipole moments, recent studies have been concentrated on such species. The Coulomb-Born

approximation has been applied by Boikova and Obédkov (1968) to HeH^+ and by Chu and Dalgarno (1974) to CH^+ . For electron-polar ion collisions the charge-dipole interaction is the one having the longest range, and it dominates over all other anisotropic interactions. Hence recent researches have been mainly concentrated on transitions through a charge-dipole interaction potential only, though both dipole and quadrupole interactions have been considered by Ray and Barua (1975).

Dickinson and Muñoz (1977) have shown that semiclassical time-dependent first-order perturbation theory gives results which are in excellent agreement with the more tedious Coulomb-Born calculations. Later, calculations of Strekalov (1979) showed that a higher order semiclassical calculation gives essentially the same result as the first order approximation for CH^+ .

Thus it is clear that in the case of electron-molecular ion collisions, the $\Delta j = 1$ transition cross sections due to the charge-dipole interaction can be calculated accurately from a semiclassical first order perturbation theory. A simple approximation like this excludes exchange and polarization phenomena and ignores competing channels such as dissociative recombination, but is adequate to estimate the effect of rotational excitations. However when the incident electron penetrates the charge cloud of the molecule, the charge-dipole potential is no longer valid. In this region strongly repulsive central and noncentral forces arise. It has not yet been possible to estimate these forces quantitatively. Dickinson and Muñoz (1977) have, however, put forward arguments to show that the net effect of the penetrating trajectories would be an enhancement of the $\Delta j = 1$ cross sections from their purely charge-dipole interaction values.

The collision rates for electron-ion collisions under interstellar conditions can be calculated by a numerical integration of the calculated cross sections. However, Flower (1979) observed that the product of energy and cross section, which is called collision strength, varies slowly with energy, and a quadratic fit of the numerically

calculated values was made from which the rate constants were derived.

In this paper we shall give a generalized expression for the rate constant which should be applicable to all systems and which follows naturally from the first order perturbation theory expression for the probabilities. After we completed our work, we came to know that Dickinson and Flower (1980) have found a similar formula for the dipolar rate coefficients. We shall discuss their results and will show that the two expressions give identical results at low temperatures—and even at high temperatures the two expressions give results which are very close.

We have also estimated the possible effect of the short-range potential on the $\Delta j = 1$ transition rate by following the method of Dickinson and Muñoz (1977). The astrophysical importance of electron-ion collisions in relation to H_2 -ion collisions has been assessed. We shall finally discuss the difficulties in applying this method to the calculation of $\Delta j = 2$ rates in which the short-range effects are likely to be much more important.

II. METHOD

a) Dipole Transition Rate

The expression for the symmetrized transition probability $P(j \rightarrow j \pm 1)$ of a linear dipolar molecular ion arising from the charge-dipole interaction may be written as

$$P = \frac{2j' + 1}{3} \begin{pmatrix} j & j' & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 \frac{v_f}{v_i} D^2 E [I_{11}^2 + I_{1-1}^2]. \quad (1)$$

Here E is the geometric mean of the initial and final kinetic energy, v_i and v_f are respectively the initial and final relative velocities, and D is the dipole moment, all in atomic units. $I_{1\mu}$ are the integrals defined by Dickinson and Muñoz (1977) and are similar to those arising in the problem of Coulomb excitation of nuclei (Alder *et al.* 1956). These are functions of the scattering angle ϑ and another parameter ξ given by

$$\xi = \frac{|\Delta E|}{(2E)^{3/2}}, \quad (2)$$

ΔE being the translation to rotation energy transfer in atomic units.

The total cross section for the transition $j \rightarrow j' = j \pm 1$ is given by

$$\sigma(j \rightarrow j') = \int P(j \rightarrow j') d\sigma_R, \quad (3)$$

where $d\sigma_R$ is the Rutherford cross section for scattering through an angle between ϑ and $\vartheta + d\vartheta$.

Thus the cross section for $\Delta j = \pm 1$ can be written as

$$\sigma = CJ(\xi), \quad (4)$$

where

$$C = \frac{\pi D^2 v_f}{24E v_i} \frac{j_{>}}{2j+1}, \quad (5)$$

and

$$J(\xi) = \int_0^\pi [I_{11}^2 + I_{1-1}^2] \sin^{-4} \frac{\vartheta}{2} \sin \vartheta d\vartheta, \quad (6)$$

$j_{>}$ is the greater of the initial and final rotational quantum numbers.

The function $J(\xi)$ can be obtained by numerically performing the I integration and then integrating over ϑ . However, comparing the integrals with similar integrals $I_{1\mu}^r$ arising in a repulsive potential, it is seen that $I_{1\mu} = e^{\pi\xi} I_{1\mu}^r$. The $I_{1\mu}^r$ integrals have been tabulated by Alder and Winther (1956) for low values of ξ and can be used in our calculations. For high values of ξ we have calculated the corresponding functions $J(\xi)$ from the table of excitation cross sections of CH^+ given by Dickinson and Muñoz (1977). These are very close to the Coulomb-Born cross sections obtained by Chu and Dalgarno (1974).

We have made a least squares fit to the function $J(\xi)$. The functional form was taken as

$$J(\xi) = A + \frac{A_1}{(\xi)^{1/2}} + \frac{A_2}{(\xi)}. \quad (7)$$

It was found that the fit reproduced all the CH^+ cross sections calculated from first order perturbation theory to within 2.5%. Also the Coulomb-Born cross section at 1.5 times the threshold energy for CH^+ was reproduced to within 10%. The constant A is determined from the low energy limit ($\xi \rightarrow \infty$) of the cross section obtained from the limiting form of the complex Hankel function (Dickinson and Muñoz 1977). When all quantities are measured in atomic units, the constants are $A = 58.04$, $A_1 = 13.10$, $A_2 = -0.3048$.

The rate constant at temperature T is $\langle \sigma v_i \rangle$, the average being over a Maxwellian distribution of velocities of the incident electrons. The integration can be performed by expressing the incident energy as $E \pm \Delta E/2$, or, in other words, by replacing the geometric mean of the incident and final energies by the arithmetic mean. The expression for the rate constant using (7) is

$$K(j \rightarrow j \pm 1, T) = \frac{7.194 \times 10^{-7}}{(T)^{1/2}} D^2 \frac{j_{>}}{2j+1} e^{\mp \Delta E/2kT} \\ \times \left[58.04 e^{-|\Delta E|/2kT} + \frac{1.521 \times 10^{-3}}{|\Delta E|^{1/2}} (T)^{0.75} \right. \\ \left. - \frac{6.47 \times 10^{-9}}{|\Delta E|} (T)^{1.5} \right], \quad (8)$$

where ΔE is in atomic units and the final rate coefficient is obtained in $\text{cm}^3 \text{s}^{-1}$. This formula automatically satisfies the principle of detailed balance.

b) Contribution of the Short Range Potential

Dickinson and Muñoz (1977) derived an expression for the contribution of trajectories penetrating the electron charge cloud to the cross section from an assumed empirical form of the probability versus impact parameter curve in this region. The expression contains a

parameter η , which is the maximum value of the unsymmetrized probability of transition attained for penetrating collisions.

Assuming $1/E \gg r_c$, the dimension of the charge cloud, in atomic units (an approximation which is valid for all energies of interest even at the highest temperature), we can derive an expression for the contribution of the penetrating trajectories to the rate coefficient

$$K^{SR}(j \rightarrow j+1, T) = 1.44 \times 10^{-6} \frac{e^{-\Delta E/kT}}{(T)^{1/2}} \times \eta [R_a(1 + 3R_a) - 8(1 + R_a)^{-1}] \quad (9)$$

Here $R_a = (2r_c)^{1/2}$, r_c being taken as roughly twice the internuclear distance R_c for a diatomic molecular ion. For a linear polyatomic molecular ion R_c is the distance between two extreme nuclei.

Estimation of η is not possible without further quantitative information about the nature and strength of short range interactions. However, some general conclusions about η can be made in the following manner. For low impact parameters where the trajectories penetrate the molecular charge cloud all the collisions are sudden. For an anisotropic potential with only P_1 angular dependence, the maximum allowed value of $|\Delta j|$ does not exceed

$$V_i = \int_{-\infty}^{\infty} V(r, t) dt.$$

The maximum transition probability thus should be inversely proportional to V_i measured in atomic units. We have assumed that the maximum is attained for an impact parameter $b_2/2$, where b_2 is the impact parameter for which trajectories just enter the charge cloud. For sudden collisions, V_i can be roughly approximated as $V'b/v$, V' being the magnitude of the anisotropic potential at the turning point at an impact parameter b . If we assume that the isotropic and anisotropic potentials are of the same order of magnitude at the turning point, then $V_i \sim E_i b_2 / (2v)$. This does not exceed unity by a large amount, and thus even at low impact parameters the higher order transitions from a pure P_1 potential will not be important. However, inside the molecular charge cloud the anisotropic potential deviates considerably from the pure P_1 form. Because of the coupling of different terms of the Legendre expansion of the anisotropic potential, η decreases. If terms up to P_n are significant in the region $b = 0$ to b_2 , giving transitions up to $\Delta j = n$, from statistical considerations, the average of the transition probabilities may be assumed to be only $1/n$ for sudden collisions. Thus for $\Delta j = \pm 1$ we take $\eta = n^{-1} j_+ / (2j + 1)$. The j -dependent factor arises from the dynamics of sudden collisions in which the $\Delta j = +1$ and $\Delta j = -1$ transition probabilities are related through the corresponding $3-j$ coefficients for a pure P_1 potential.

If we choose a rigid ellipsoid model for the short range interaction having a and b as the semimajor and semiminor axes, respectively, then only terms up to P_n will be

significant, n being given by a/b . For the molecular ions we make the rough estimate $n \approx r(\text{atom})/R_c$, where $r(\text{atom})$ is the mean atomic radius of the isolated atoms comprising the molecular ion. These values for different ions have been obtained from Fraga *et al.* (1976). This rough estimate gives $n = 2$ for CH^+ , 4 for N_2H^+ , and 5 for HCO^+ . For the polyatomic ions, internuclear separations calculated by Haese and Woods (1979) were used.

The pure dipole cross sections are estimated by integrating over all impact parameters. However, since the dipolar contribution from the region $b = 0$ to b_2 is generally a small part of the total dipole cross section and also in view of the fact that there are many other sources of uncertainty in the evaluation of short range cross sections, the two rates may be simply added to yield the total $\Delta j = \pm 1$ transition rate. The downward rate may be obtained from the principle of detailed balance.

c) $\Delta j = 2$ Transitions

For molecular ions with finite quadrupole moments there will be a long range charge-quadrupole interaction causing $|\Delta j| = 2$ transitions. This has a much shorter range than the charge dipole interaction, and the first order transition probability exceeds unity for close collisions. At this range of impact parameters higher order processes will start contributing. Though the quadrupole moments of the relevant molecular ions are not known, it is expected that the strong coupling region will be dominant over the first order perturbation region as far as $|\Delta j| = 2$ transitions are concerned. The short range interactions also start becoming important in this region. Under the circumstances, the best that can be done is to assume a contribution of the strong coupling region to the $|\Delta j| = 2$ transition similar to the $|\Delta j| = 1$ transition. This contribution will generally dominate over the first order cross section obtained from the weak coupling region. Hence we may use the same equation (9) for the $\Delta j = 2$ rate. In this the j -dependent factor in η should be given by $3(j+1)(j+2)/2(2j+3)(2j+1)$. This factor follows from the geometrical factor in sudden collision dynamics.

III. RESULTS AND DISCUSSION

For pure dipole transitions, the trajectory integrals $I_{1\mu}$ in the attractive and repulsive Coulomb fields are related to each other. Indeed, the ratio of cross sections for rotational excitation of HD^+ in collision with electrons and positrons should be $e^{2\pi\xi}$. This ratio is satisfied in the calculations of Ray and Barua (1975) at all energies greater than 0.1 eV. The discrepancy of 5–12% as observed is due to the short-range cutoff used by them.

The values of molecular constants used in the calculations are shown in Table 1. The rate coefficients for CH^+ obtained by us are in good agreement with the calculations of Dickinson and Flower (1980) at low energies, which is expected since in both cases the fit is made by considering the low energy limit of the dipole cross section. However, at high energies also the fit is satisfactory, and for CH^+ the agreement is within 3% at all temperatures. For N_2H^+ , the maximum discrepancy is

TABLE 1

VALUES OF DIPOLE MOMENT μ , ROTATIONAL CONSTANT B , AND R_e (the internuclear distance) OF CH^+ , N_2H^+ , AND HCO^+

Parameter	CH^+	N_2H^+	HCO^+
R_e (Å)	1.23 ^a	2.13 ^b	2.20 ^b
μ (debye)	1.70 ^c	3.50 ^b	4.07 ^b
B (cm^{-1})	13.9 ^c	1.55 ^d	1.49 ^d

^a Huber and Herzberg 1979.^b Haese and Woods 1979.^c Chu and Dalgarno 1974.^d Turner and Thaddeus 1977.

5% at 1000 K when the same value of dipole moment is used.

The values of rate coefficients at a few representative temperatures are given in Table 2 for $0 \rightarrow 1$ and $1 \rightarrow 2$ transitions. It is seen that the maximum occurs near a temperature T such that $kT \approx 2\Delta E$ and the peak is flatter for higher ΔE transitions. The ratio of the short range contribution to the pure dipole rate will have a weak temperature dependence. The ratio is higher for molecules of larger size and lower dipole moment. Thus for $0 \rightarrow 1$ transition in HCO^+ the ratio varies from 13.5% to 8.5% while for the same transition in CH^+ the

TABLE 2

RATE COEFFICIENTS (dipolar, short-range, and total) (in $10^{-7} \text{ cm}^3 \text{ s}^{-1}$) OF CH^+ , N_2H^+ AND HCO^+ AT SOME REPRESENTATIVE TEMPERATURES IN COLLISION WITH ELECTRONS

	$T(\text{K})$				
	10	20	50	100	500
CH^+					
Dipolar	1.24	6.17	13.0	14.1	10.4
	0.02	0.57	3.82	6.05	5.64
Short range	1.22	6.33	13.3	14.0	8.62
	0.02	0.57	0.39	6.2	5.25
Total	2.46	12.5	26.3	28.1	19.0
	0.04	1.14	7.76	12.5	10.9
N_2H^+					
Dipolar	167	152	116	92.2	57.8
	70.2	78.4	68.5	54.4	34.3
Short range	36.9	32.6	23.6	17.4	8.09
	16.0	17.7	14.7	11.4	5.45
Total	204	185	140	110	65.9
	86.2	96.1	83.2	65.8	39.8
HCO^+					
Dipolar	231	207	158	125	78.8
	100	110	93.5	76.0	46.7
Short range	31.1	27.3	19.6	14.5	6.70
	13.2	14.3	11.7	7.41	4.32
Total	262	234	178	140	85.5
	113	124	105	83.4	51.0

NOTE.—The upper value is the rate coefficient for the $0 \rightarrow 1$ transition and the lower value is that for the $1 \rightarrow 2$ transition.

ratio lies between 98% and 83%. for $\Delta E \ll kT$ the ratio increases with increasing ΔE .

For $j = 2$ or 3 , $\Delta j = 1$, the condition $\Delta E \ll kT$ is well satisfied for N_2H^+ and HCO^+ near or above 30 K and thus the relative short range contributions to these transitions is somewhat greater compared to $0 \rightarrow 1$ transition. This argument is not valid for larger j and at very low temperatures, and the relative contribution of the short range interactions to the rate coefficient may decrease with increasing j .

Since the dipole moment and internuclear distance of N_2H^+ are similar to those of the neutral molecule HCN, it is interesting to compare the effect of electron collisions on ionic and neutral molecular species by comparing the rate coefficients for neutral molecules and molecular ions. The rate coefficients for e -HCN collisions have been calculated by Saha *et al.* (1980). The HCN rates are lower than those of N_2H^+ by only about 10–15% at 100 K but are smaller by a factor of 5 to 6 at low temperatures (5–10 K). This is due to the fact that at low energies the effect of the attractive Coulomb interaction for electron-ion collisions is more important. The effect of adiabaticity of the collisions is decreased by the attractive field; therefore the effect will be greater for more adiabatic collisions, i.e., for higher energy transfers. Thus the difference between the HCN and the N_2H^+ rates increases with increasing initial j .

Now we compare the effectiveness of electrons and molecular hydrogen in exciting the rotational levels of molecular ions. This will enable us to determine the range of electron concentrations in which electron-ion collisions should be considered in addition to H_2 -ion collisions to determine the population distribution of the rotational levels of the ion in the interstellar cloud. We consider the case of N_2H^+ since a close-coupling calculation has been done for the rate coefficient of H_2 - N_2H^+ collisions (Green 1975) from an ab initio potential energy surface. It is seen that the ratio of the two rates changes from 1.4×10^5 at 5 K to 6.7×10^4 at 40 K for the $0 \rightarrow 1$ transition in N_2H^+ . For the $1 \rightarrow 2$ transition the ratios are 9.2×10^4 and 4.4×10^4 , respectively. Thus the electron-ion collisions will be significant compared to that of H_2 -ion collisions for electron concentrations of 10^{-5} times that of H_2 or larger. Similar values for neutral HCN molecule has been obtained for optically thin clouds by Dickinson *et al.* (1977) and for optically thick clouds by Bhattacharyya and Baur (1981a). The transition rates for HCO^+ are somewhat higher than for N_2H^+ but the relative importance of the electron and hydrogen collisions should be of the same order of magnitude.

An estimate of the ratio of electron to H_2 abundance in some interstellar clouds has been given by Wootten, Snell, and Glassgold (1979) from a model of gas-phase ion-molecule reactions. Their method can deal with both dense cool clouds and the warmer ones. On the basis of the observational data available on H_2 , HCO^+ , and CO densities they deduce that the relative electron densities are less than 10^{-7} for a large number of clouds where these ions are observed. These values are consistent with those obtained from a study of the deuteration of HCO^+

(Watson, Snyder, and Hollis 1978), applicable only to cold clouds. Thus for those clouds listed, the effect of electron collisions on excitation of ionic rotational lines will not be important. However, at the center of Orion A where these molecular lines have been observed (Turner and Thaddeus 1977), the electron concentration was estimated to be much higher. Near the center a mean value of 8×10^{-5} for the electron- H_2 ratio was established by Bhattacharyya and Barua (1981b) from considerations of HCN and ^{13}CO excitation. A value greater than 5×10^{-4} for Orion clouds was also suggested from a similar argument by Phillips and Huggins (1977). Thus at the center of the Orion A cloud where both HCO^+ and N_2H^+ have relatively high brightness temperatures, the

effect of electron ion collisions seems to be important.

For $\text{H}_2\text{-N}_2\text{H}^+$ close-coupling calculations the $|\Delta j| = 2$ rates are almost as important as the $|\Delta j| = 1$ rates. They differ by a factor of 2–3. Our estimates of the $|\Delta j| = 2$ rates are very approximate, and numerical values cannot be taken too seriously. However, in our model the $\Delta j = 2$ rates come out to be lower than the $\Delta j = 1$ rates by a factor 6–7 for initial $j = 0$.

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