Eur. Phys. J. D **46**, 83–87 (2008)
DOI: 10.1140/epjd/e2007-00313-4

THE EUROPEAN PHYSICAL JOURNAL D

Zeeman relaxation of N_2^+ ($^2\Sigma^+$) in collisions with ^{3}He and ^{4}He

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Received 5 July 2007 / Received in final form 18 September 2007

Abstract. We compare the cross sections for the transitions changing the projection of the total angular momentum of $N_2^+(^2\Sigma)$ in collisions with ^{3}He and ^{4}He at very low collision energy. The fundamental states of the two nuclear spin isomers of N_2^+ are considered as well as the two fine structure levels of the first excited para level $N = 2$. It is shown that the two fundamental states of the two nuclear spin isomers behave differently. For the fundamental para level $N = 0$ of N_2^+, the projection changing cross section is always negligible compared to the elastic one for both He isotopes. For the fundamental ortho level $N = 1$ of N_2^+, the spin-rotation interaction couples the different spin levels directly so the spin relaxation becomes a first order process. The associated resonances increase the projection changing cross section which remains smaller but becomes comparable with the elastic one. This is in contrast with the excited rotational levels of N_2^+, which for the rotational deactivation and elastic channels are found to be equal around the resonances for the collisions involving ^{3}He. These two channels are always larger than the projection changing one. We also find that, for transitions involving the fundamental rotational state, the domain of validity of the threshold laws discussed by Krems and Dalgarno [Phys. Rev. A **67**, 050704 (2003)] for a potential decreasing faster than $1/r2$ is shortened, due to the long range charge induced dipole potential. This effect is illustrated for the collisions of ^{3}He with the fundamental para state of N_2^+.

PACS. 34.50.Ez Rotational and vibrational energy transfer – 34.50.Pi State-to-state scattering analyses

1 Introduction

The recent experimental progresses achieved in the production of cold neutral [1–4] and ionic [5,6] molecules have stimulated many theoretical studies [7,8] dedicated to the understanding of collisional energy transfer at very low temperature and to the optimisation of both the cooling process and the trapping techniques. These progresses also open the way to controlling the outcome of the collisions using electric [9,10] or magnetic fields [11–14]. Cold molecules are being produced by a wide variety of techniques. Among them, the buffer gas loading method, which is the most universal, is based on energy equilibration of molecules in elastic collisions with cold buffer gas atoms and subsequent trapping of the cooled molecules. Magnetic trapping is currently used to trap neutral paramagnetic molecules. In order to be magnetically trapped, paramagnetic molecules must be stable against reorientation of their magnetic moments in collisions with other trapped molecules or sympathetic atomic coolant. The reorientation of the molecules called Zeeman relaxation is of critical importance as it leads to trap loss and was consequently the subject of several recent studies [15–17]. The most comprehensive one was dedicated to the He CaH collisions which for experimental data are available in the ultra cold regime. Krems et al. [18] studied both the effect of the spin rotation interaction and the Zeeman relaxation in the fundamental rotational level for this system. They concluded that the collision-induced spin-flip transitions will be least efficient for diatomic molecules with large rotational constants and small spin rotation constants γ, interacting with a collision partner through a weakly anisotropic potential. They also suggested that they may be enhanced if the atom-molecule interaction is strongly attractive with a deep potential well. In a previous paper [19] we investigated the effect of the spin-rotation interaction for a collision involving an ionic molecule: the He + N_2^+ collision. This system has a deep well and a strongly attractive long range charge induced dipole potential. It is furthermore comparable to the He + CaH collision as the electronic states $^2\Sigma$ of the two diatomic molecules are the same and the ratio B/γ of $N_2^+(^2\Sigma)$ is twice as large as for CaH. Another great interest of this system is that the two nuclear spin isomers of N_2^+ can be studied separately both experimentally and theoretically and that the spacing between the fundamental and the first excited rotational levels are different for the two species. Krems et al. showed that the mechanism for spin flipping transitions in the fundamental rotational state ($N = 0$) are induced through the coupling to the excited rotational levels. On the other side Zeeman relaxation of excited rotational levels follows

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$$\sigma_{jm_j\varepsilon\to j'm_{j'}\varepsilon'} = \frac{\pi}{k_j^2}\sum_J\sum_{J'}\sum_{l=\left|J_-j\right|}\sum_{m_l}\sum_{l'=|J'-j'|}\sum_{m_{l'}}(2J_+1)\,(2J'_+1)\,(-)^{2\left[J+J'+m_j+m_l+m_{j'}+m_{l'}\right]}T^{J*}_{jl\varepsilon;j'l'\varepsilon'}T^{J'}_{jl\varepsilon;j'l'\varepsilon'}$$

$$\times\begin{pmatrix} j & l & J \\ m_j & m_l & m_j+m_l\end{pmatrix}\begin{pmatrix} j & l & J' \\ m_j & m_l & m_j+m_l\end{pmatrix}\begin{pmatrix} j' & l' & J \\ m_{j'} & m_{l'} & m_{j'}+m_{l'}\end{pmatrix}\begin{pmatrix} j' & l' & J' \\ m_{j'} & m_{l'} & m_{j'}+m_{l'}\end{pmatrix}. \quad (1)$$

a direct mechanism. In the case of N_2^+, the Zeeman relaxation of the two lowest-lying ortho ($N = 1$) and para ($N = 0$) states of N_2^+ are then expected to behave in a very different way. In the present work we study the collision induced Zeeman relaxation for this system. We focus first our attention on the lowest-lying para ($N = 0$) and ortho ($N = 1$) states of N_2^+ in collisions with both ^{3}He and ^{4}He. We will also examine the case of the excited rotational states and use the results obtained in our previous work dedicated to the spin rotation interaction for this collision. The paper is organised as follows. In Section 2 we shortly remind the main features of the method and the parameters used to make the scattering calculations. Our results are then presented and analysed in Section 3.

2 Theory and parameter calculations

The calculations were performed using the potential energy surface and the dynamics code presented in our previous papers [19,20]. Spin rotation was taken fully into account and the cross sections for projection-flipping cross sections for a given transition $j\ m_j\ \varepsilon \to j\ m_{j'}\ \varepsilon'$ were evaluated from the T matrix using the usual formula [18]

see equation (1) above.

In this expression j, l and J are respectively the initial values of the rotational, relative and total angular momentum quantum numbers. ε is the initial value of the parity, m_j and m_l denote the space fixed projections of j, l respectively. The primes designate the values of the different quantum numbers after collision. The results of these calculations were checked by summing over $m_{j'}$ and averaging over m_j. The resulting inelastic cross sections were found to be identical to those presented in our previous paper. The cross sections were calculated in the ultra cold energy regime between 0.0001 and 1 cm^{-1} which covers the region of action of the spin rotation interaction. More details about the calculation parameters are given in our previous work [19,20].

3 Results and analysis

3.1 Spin-flipping transitions for the fundamental para and ortho rotational levels of N_2^+

From now, we will always choose the positive projection m_j as an initial projection quantum number for the various transitions. Indeed, transitions from the corresponding negative projection will have the same absolute value of $\Delta m = (m_{j'} - m_j)$ and the associated cross sections are thus identical by symmetry of the Wigner $3-j$ coefficients entering in expression (1) of the projection changing cross section. Figures 1a and 1b show the cross section for the spin-flipping transition ($m_j = +1/2 \to m_{j'} = -1/2$) from the fundamental para rotational level ($N = 0$, $j = 1/2$, $\varepsilon = -$) of N_2^+ together with the elastic cross section which conserves the value of m_j respectively in collisions with ^{3}He and ^{4}He. On both figures we can first notice that there are no resonances in the energy regime where the spin rotation is important (below 0.1 cm^{-1}). As noticed by Krems et al. [18] the spin rotation interaction in the fundamental rotational state $N = 0$ is induced through the coupling to the excited rotational levels. The rotational spacing between the fundamental and the first rotationally

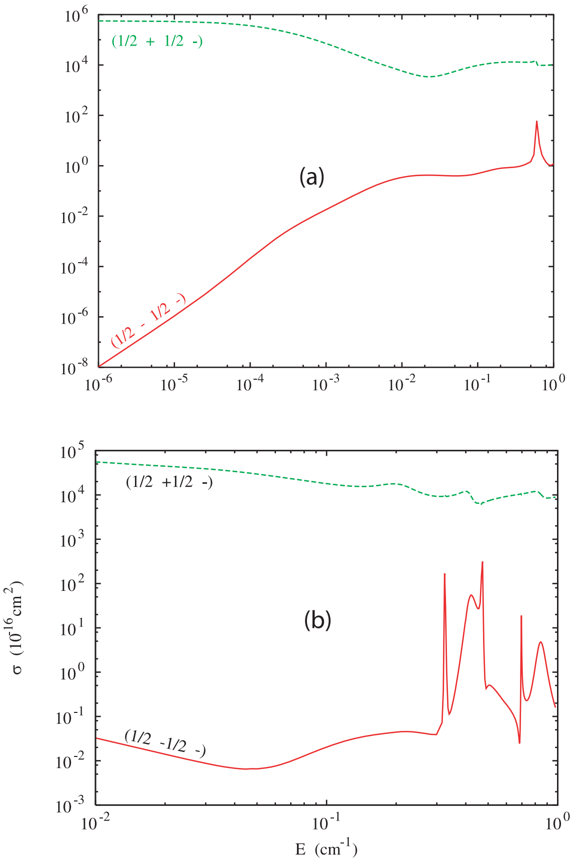

Fig. 1. (Color online) Comparison of the cross sections for spin-flipping and elastic scattering transitions of $N_2^+(N = 0, j = 1/2, m = 1/2, \varepsilon = -)$ in collisions with ^{3}He (a) and ^{4}He (b).

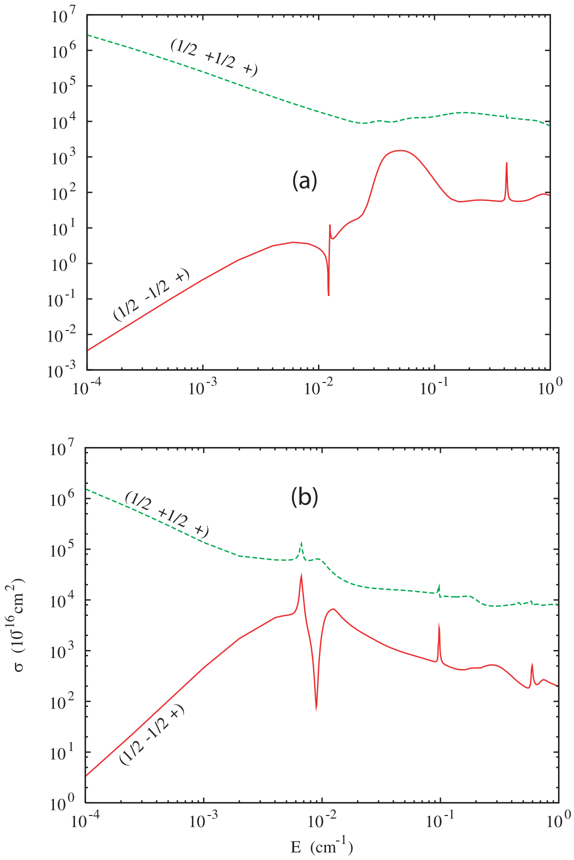

Fig. 2. (Color online) Comparison of the cross sections for spin-flipping and elastic scattering transitions of $N_2^+(N = 1, j = 1/2, m = 1/2, \varepsilon = +)$ in collisions with ^{3}He (a) and ^{4}He (b).

excited states are respectively 8.54, 11.59 and 19.32 cm^{-1} for CaH$(^2\Sigma)$ and the para and ortho isomers of $N_2^+(^2\Sigma)$ The rotational spacing for para N_2^+ is then larger than its CaH counterpart. Since the spin rotation constant is furthermore smaller for N_2^+ than for CaH it is then not surprising to not see any effect at very low energy. Despite the strongly attractive long range potential and the deep well the spin flipping cross section is smaller by four orders of magnitude than the elastic cross section and is then negligible for this system. There is another striking difference between these two systems which is due to the difference between the long ranges potentials associated with these two collisions. This effect is illustrated in Figure 1a for the ^{3}He–N_2^+ collisions where a low energy minimum of the elastic cross section is clearly apparent which was absent in the case of the He-CaH collisions. This is a Ramsauer Townsend minimum associated with the long range charge induced dipole potential. In a recent work Krems and Dalgarno [21] showed that the threshold laws for the projection changing transitions in collisions of paramagnetic molecules with structureless targets have the form $v^{2\Delta m}$ when Δm is even and $v^{(2\Delta m+1)}$ when Δm is odd. If we compare in Figure 1a our results with those obtained by Krems et al. [18] for the He-CaH collision we see that the presence of the charge induced dipole potential shortens the domain of validity of the threshold law for the projection changing transitions to the very low energy range. The cross section for the spin-flipping transition together with the elastic cross section which conserves the value of m_j are also compared for the fundamental $(N = 1, j = 1/2, \varepsilon = +)$ ortho rotational state of N_2^+ in Figures 2a and 2b again respectively in collisions with ^{3}He and ^{4}He. The spin rotation interaction acts directly on this level and we can see clearly a first resonance which energy position $(1.25 \times 10^{-2}$ cm$^{-1})$ corresponds to the doublet shifting of the $N = 1$ level. We showed in our previous work that it is a Feshbach resonance associated with the opening of the negative parity level of the $N = 1$ doublet. This resonance can be seen on the two figures associated with the two isotope of He but is much stronger for the collisions involving ^{4}He. The resonances associated with the spin rotation interaction and consequently the spin flipping cross sections are then increased with the increase of the relative mass. The cross section for the projection changing transition is much bigger than for the fundamental rotational para state of N_2^+ and increases (in the case of ^{4}He) by 5 orders of magnitude around the resonance. Conversely to the cross section associated with the $N = 0$, the domain of energy where the spin flipping cross section decreases as E_{col}^2 with decreasing collision energy E_{col} as predicted by Krems and Dalgarno [21] extends up to 2×10^{-3} cm^{-1}. When the spin rotation is non zero, there is no longer restriction of the domain of validity of the threshold law.

A similar comparison is presented in Figures 3a and 3b for the negative parity $(N = 1, j = 3/2, \varepsilon = -)$ ortho rotational state of N_2^+ again respectively in collisions with ^{3}He and ^{4}He. For the whole range of energy considered in this figure, the elastic cross sections again are the largest. There are no Feshbach resonances to be seen as they can only be found for transitions out of the positive parity state as analysed in our previous work. The fine structure relaxation towards the fundamental ortho rotational state which is now open dominates the projection changing channel at very low energy for both He isotopes. When the energy increases these two channels are of the same order of magnitude.

3.2 Transitions arising from excited rotational levels of N_2^+

3.2.1 Transitions from j = 3/2

We now consider the transitions arising from the first rotationally excited para levels $N = 2$ of N_2^+. Figure 4 shows the cross section for the projection changing from the $(N = 2, j = 3/2, m = 3/2, \varepsilon = +)$ level of N_2^+ together with the elastic and the rotational deactivation cross section respectively in collisions with ^{3}He and ^{4}He. If, for the whole range of energy considered in this figure and for the two He isotopes, the elastic cross sections remain the largest, the rotational deactivation towards the fundamental $(N = 0, j = 1/2, e = 1)$ state is now the second largest before those associated with the projection

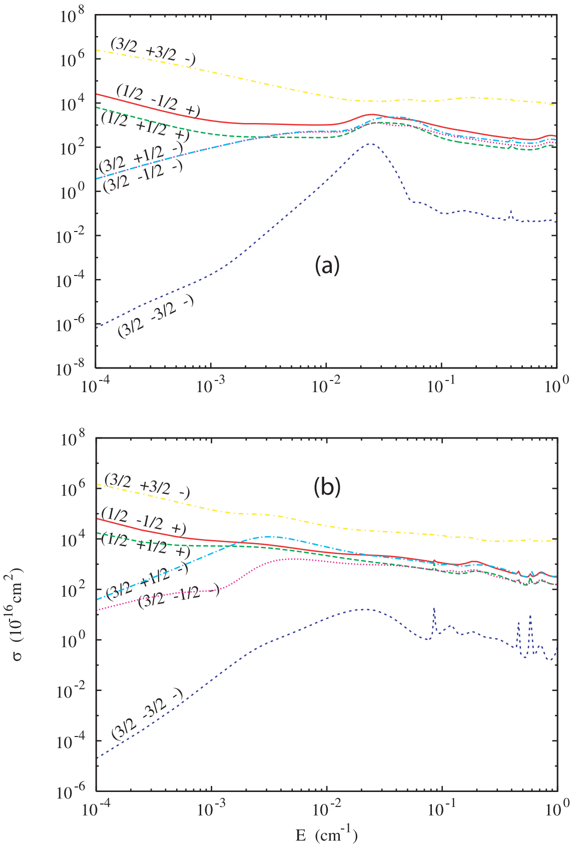

Fig. 3. (Color online) Comparison of the cross sections for projection changing, elastic scattering and fine structure deactivation transitions of N_2^+($N = 1$, $j = 3/2$, $m = 3/2$, $\varepsilon = -$) in collisions with ^{3}He (a) and ^{4}He (b).

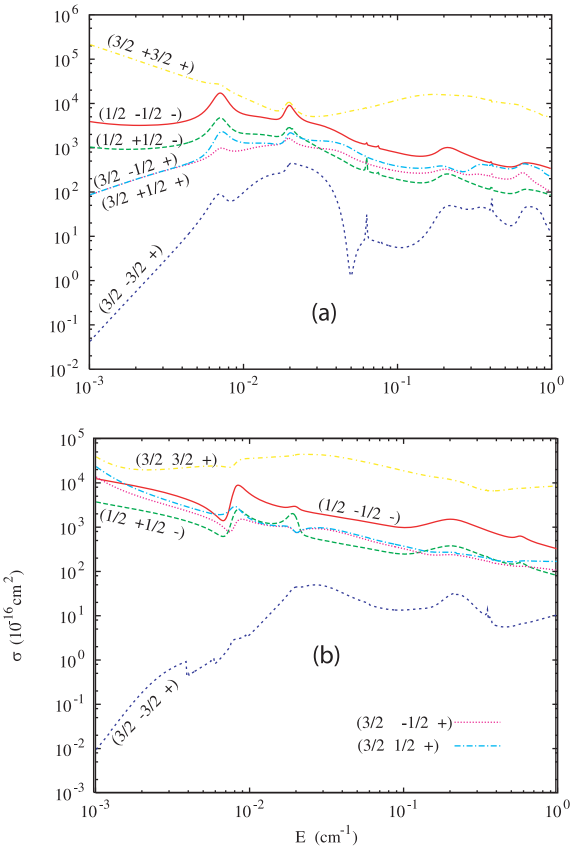

Fig. 4. (Color online) Comparison of the cross sections for projection changing, elastic scattering and rotational deactivation transitions of N_2^+($N = 2$, $j = 3/2$, $m = 3/2$, $\varepsilon = +$) in collisions with ^{3}He (a) and ^{4}He (b).

changing channels. The two resonances appearing respectively at the collision energies 0.007 cm^{-1} and 0.02 cm^{-1} have been analysed in our previous work. The second one is a Feshbach resonance due to the opening of the ($N = 2$, $j = 5/2$, $\varepsilon = -$) channel whereas the first one is a shape resonance. Around these two resonances the cross sections associated with all the other channels increase also significantly. There is however a striking difference between the collisions involving the ^{3}He and ^{4}He isotopes. The rotational deactivation and elastic cross sections are equal around the Feshbach resonance on the curve associated with the collisions involving ^{3}He whereas the rotational deactivation channel remains smaller than the elastic one for the collisions involving ^{4}He. In average, and particularly at low collision energy, the magnitude of projection changing cross sections decrease when $|\Delta m|$ increases. The cross section associated with the maximum value of Δm_j is always the lowest in magnitude. The same trend could already be seen in Figure 3.

3.2.2 Transitions from j = 5/2

A similar comparison is presented in Figure 5 for the transitions arising from the negative parity para level ($N = 2$, $j = 5/2$, $m = 5/2$, $\varepsilon = -$) of N_2^+. The elastic cross sections again are the largest and again are equal to the rotational deactivation around the resonance only for the collisions involving ^{3}He. Apart in a narrow interval at very low energy, the fine structure relaxation towards the $N = 2$ ortho rotational state remains smaller than the projection changing channel for both He isotopes. These figures also show an illustration of the threshold laws for the collisional reorientation cross sections obtained by Krems and Dalgarno [21].

4 Conclusion

The Zeeman relaxation of the two lowest-lying ortho ($N = 1$) and para ($N = 0$) states of a homonuclear molecule like N_2^+ are found to behave very differently. This is a consequence of the fact that the mechanism for spin flipping transitions in the fundamental rotational state ($N = 0$) are induced through the coupling to the excited rotational levels whereas Zeeman relaxation of the $N = 1$ levels follow a direct mechanism. For the fundamental para level $N = 0$ of N_2^+, the projection changing cross section is always negligible compared to the elastic one for both He isotopes. This is due to the fact that the rotational spacing for para N_2^+ is quite large

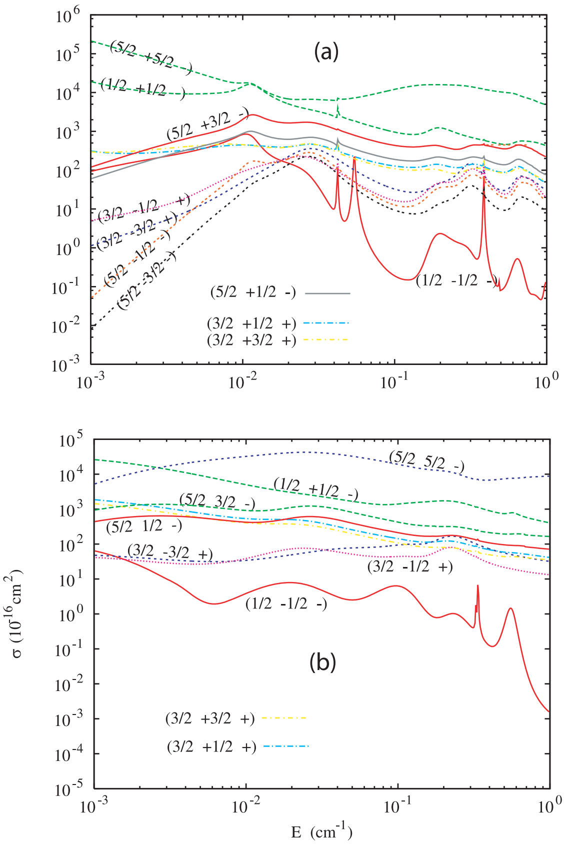

Fig. 5. (Color online) Comparison of the cross sections for projection changing, elastic scattering, fine structure and rotational deactivation transitions of $N_2^+(N = 2, j = 5/2, m = 5/2, \varepsilon = -)$ in collisions with ^{3}He (a) and ^{4}He (b).

and that the spin rotation constant is small. We also find for the fundamental para state of N_2^+, that the long range charge induced dipole potential shortens the domain of validity of the threshold laws for the collisional projection changing transitions obtained by Krems and Dalgarno. For the fundamental ortho level $N = 1$ of N_2^+, the spin rotation interactions creates resonances which increase the projection changing cross section. They however remain smaller by a factor of five than the elastic one. This is in contrast with the excited rotational levels of N_2^+, which for the rotational deactivation and elastic channels are found to be equal around the resonances for the collisions involving ^{3}He. These two channels are always larger than the projection changing one.

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