The influence of isotope substitution of neon atom on the integral cross sections of rotational excitation in Ne–Na_2 collisions*

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This paper applies the multiple ellipsoid model to the ^{16}Ne (^{20}Ne, ^{28}Ne, ^{34}Ne)–Na_2 collision systems, and calculates integral cross sections for rotational excitation at the incident energy of 190 meV. It can be seen that the accuracy of the integral cross sections can be improved by increasing the number of equipotential ellipsoid surfaces. Moreover, by analysing the differences of these integral cross sections, it obtains the change rules of the integral cross sections with the increase of rotational angular quantum number J', and with the change of the mass of isotope substitution neon atom. Finally, the contribution of different regions of the potential to inelastic cross sections for ^{20}Ne–Na_2 collision system is investigated at relative incident energy of 190 meV.

Keywords: multiple hard-ellipsoid model, Ne isotopic atoms, integral cross sections, ellipsoid equipotential surface

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1. Introduction

The inelastic vibrational scattering of an atom by a diatomic molecule is of both theoretical and practical importance.[1−4] The inelastic scattering of an atom by a diatomic molecule is of great importance in ultrasonic dispersion, the cooling of interstellar matter, resonance fluorescence, predicting the laser action and thermal scattering in mixture gases etc.[5−8] Collision-induced rotational state change, or rotational transfer, is very important in chemistry and physics since it provides a fast and efficient mechanism for equilibration in the gas phase.[9] From the inelastic scattering cross sections, we can learn the detail of energy transfer in atom–molecule collisions,[10,11] the experimental measure and theoretical calculation of excited cross section for rotational transition can provide some information of the intermolecular interaction potential.[8,12−16] On the other hand, the development of the experimental technique in the crossed molecular beam requires a deeper theoretical research into atom–molecule collisions.[17]

Nowadays, the study of the cold molecule has become more and more important and it has attracted people's attention.[18−23] Alkali metal can be cooled to nK by some certain techniques, and then ultra cold molecules can be produced. With the development of ultra cold molecule physics, some areas have developed rapidly, such as the research of characteristics of time and space in Einstein's relativity, precision measurement of Bose–Einstein condensate and quantum phase transitions of degenerate Fermi gas, development of quantum computer etc. Therefore, for the research and application of cold molecule, it is important to investigate the property of Na_2 molecule, and we will investigate integral cross sections of rotational excitation in Ne–Na_2 collisions.

The collision between a neon atom and a sodium molecule is one of the important molecular scattering systems and it has received much attention. For example, Jones *et al.*[8] published theoretical interaction potential for the Ne–Na_2 system, and calculated infinite order sudden (IOS) approximation integral cross sections and differential cross sections. Osborne and McCaffery[24] applied the single hard-

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shell model to ^{20}Ne–Na_2 collision system and calculated the rate constants of rotational excitation. Clare *et al.*[25] applied the multiple hard-ellipsoid model to ^{20}Ne–Na_2 collision system and calculated integral cross sections of rotational excitation. The multiple hard-ellipsoid model is more reasonable than the single hard-shell model, it is designed to be used for 'soft' and anisotropic repulsive intermolecular potentials for which single hard-shape models fail to reproduce the integral cross sections.[7] Moreover, the multiple hard-ellipsoid model provides a simple physical origin to the collisional phenomena in atom–molecule collisions which have been proved difficult to reveal by other methods. Therefore, it is a simple and effective method that can be used to calculate integral cross sections of rotational excitation in Ne–Na_2 collisions.[24−26]

Since Ne has several isotopes, in order to obtain the magnitude of the isotope effect in practical systems and to determine the accuracy in the measurement which is influenced by this effect, the effect of isotopic substitution must be realized. Although some research results have been reported for ^{20}Ne–Na_2 collisions, so far as we know, few have been reported on the influence of isotope substitution in Ne–Na_2 collisions and few data are available. In this paper, theoretical calculations of the integral cross sections for rotational excitation in ^{16}Ne (^{20}Ne, ^{28}Ne, ^{34}Ne)–Na_2 collisions are presented.

2. Theoretical methods[7]

The interaction between Ne atom and Na_2 molecule is approximated by a series of hard ellipsoids, with each shell representing an equipotential surface. The Ne atom is likely to be scattered from one of the ellipsoid equipotential surfaces. Generally, we expect that a larger number of ellipsoids are required to model the cross sections as the collision energy E_r is increased. The centre of each ellipsoid corresponds to the centre-of-mass of the Na_2 molecule, which is assumed to be a rigid rotor having bond length, r_e = 3.1 Å (1 Å=0.1 nm). In the molecule-fixed (X, Y, Z) coordinate system, the i-th ellipsoid surface can be written as

$$\frac{X^2}{A_i^2}+\frac{Y^2}{B_i^2}+\frac{Z^2}{B_i^2}=1, \tag{1}$$

where A_i and B_i are the semi-major and semi-minor axes of the i-th ellipsoid, which are extracted from the potential-energy function V by finding the value of R at a specified contour energy for $\alpha = 0°$ and $\alpha = 90°$ respectively, R is the distance between Ne atom and the centre-of-mass of the Na_2 molecule, α is the orientation angle between R and the inter-nuclear axis of Na_2. The potential-energy function V corresponds to the interaction potential for the Ne–Na_2 system which is derived by Jones *et al.*[8]

$$\begin{aligned} V =\ & a_1\exp(-a_2R_1)+a_1\exp(-a_2R_2)\\ & +b_1\exp(-c_1R-d_1R^2)\mathrm{P}_0(\cos\alpha)\\ & +b_2\exp(-c_2R-d_2R^2)\\ & \times\mathrm{P}_2(\cos\alpha)+b_3\exp(-c_3R-d_3R^2)\\ & \times\mathrm{P}_4(\cos\alpha)+e(R-f)^{-6}G^2[\,g(R-h)](2), \end{aligned} \tag{2}$$

where R_1, R_2 are the distances between the Ne atom and each of the Na atoms; $\mathrm{P}_k(\cos\alpha)$ are Legendre polynomials; $G(y) = 1-(1+y+y^2/2)\exp(-y)$ is a cut-off function; the 19 parameters a_1 through h_2 are given in Ref. [8].

The assigned potential energy of the i-th ellipsoid is written as V_i, it does not correspond exactly to the intermolecular potential contour V from which the ellipsoid parameters A_i, B_i were extracted. However, the assigned energies always satisfy the condition:

$$V_{i-1} < V \le V_i. \tag{3}$$

We assume that the incoming Ne atom approaches the ellipsoid in the xOy plane, as shown in Fig. 1.

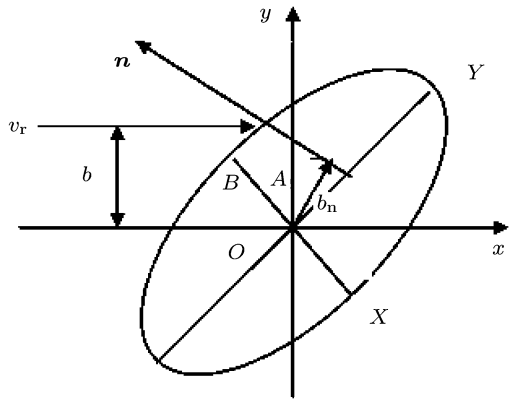


Fig. 1. The hard ellipse model that forms the basis of the multi hard-ellipsoid model. The XOY is the body-fixed coordinate frame, xOy is the space-fixed coordinates, v_r is the initial relative speed, $\boldsymbol{n}$ is the unit vector normal to the ellipsoid surface, v_n is the surface normal velocity component, b is the impact parameter, b_n is the effective impact parameter (or torque arm) about which linear momentum is converted to angular momentum.

First, we should determine whether the incident Ne atom hits the outermost equipotential surface. If there is a hit, we compare the 'normal component' of kinetic energy $E_n = \mu v_n^2/2$ with the energy assigned

to that ellipsoid surface, V_1. If $E_n \leq V_1$, then we assume that the atom is scattered impulsively by this ellipsoid; if $E_n > V_1$, then we assume that the atom does not 'feel' this equipotential surface and continues unperturbed along a straight-line path until it encounters the next ellipsoid surface or misses the molecule altogether. The process is repeated inwards until the atom is either scattered by one of the ellipsoids or flies past the molecule. Thus, each incoming atom is scattered impulsively from a single ellipsoid and its trajectory is unaltered by the outer surfaces of lower energy through which it initially passes.

We assume that the Na_2 molecule is initially non-rotating before Ne atom hits the ellipsoid equipotential surface. Then the final rotational angular momentum of the Na_2 molecule is given classically by

$$j' = \frac{2Il}{\mu b_{\rm n}^2 + I}, \tag{4}$$

where μ is the reduced mass of the Ne–Na_2 system, I is the moment of inertia of the Na_2 molecule, $l = \mu v_{\rm n} b_{\rm n}$.

The resultant rotational angular momentum is binned to obtain a final rotational quantum number according to

$$h(J'-1)/2\pi \leq j' < h(J'+1)/2\pi, \tag{5}$$

then $j' = J'h/2\pi$, J' is the final rotational angular momentum quantum number of Na_2 molecule.

To compute integral cross sections for the process $J = 0 \to J'$, we need to find the transferred rotational angular momentum for a large number of atom–diatom trajectories. We have used a Monte Carlo (MC) approach similar to that process, and MC trajectories is 10^6. According to Born–Oppenheimer approximation, the interaction potential is the same for the ^{16}Ne (^{20}Ne, ^{28}Ne, ^{34}Ne)–Na_2 systems although the masses of the isotope neon atoms are different.

The integral cross sections of rotational excitation for the process $J = 0 \to J'$ are given by

$$\sigma(J = 0 \to J') = \frac{n(J')}{N}\pi b_{\rm max}^2, \tag{6}$$

where $n(J')$ is the number of MC trajectories resulting in a final rotational quantum number J', and $b_{\rm max} = A_{\rm i\,max}$ is the maximum impact parameter.

The total collision cross section is given by

$$\sigma_{\rm tot} = \sum_{J'=2,4,6\ldots} \sigma(J = 0 \to J') = \frac{n_{\rm hits}}{N}\pi b_{\rm max}^2, \tag{7}$$

where $n_{\rm hits}$ is the total number of trajectories in which the neon atom hits the ellipsoid.

3. Results and discussion

For each α_k, the R dependence of the interaction potential is described by a cubic spline function[8] as

$$\begin{aligned} F(R,\alpha_k) &= \ln V(R,\alpha_k) \\ &= A_k^{(i)} + B_k^{(i)}(R - R_i) + C_k^{(i)}(R - R_i)^2 \\ &\quad + D_k^{(i)}(R - R_i)^3, \end{aligned}$$

where $R_i \leq R \leq R_{i+1}$, $A_k^{(i)}$, $B_k^{(i)}$, $C_k^{(i)}$, $D_k^{(i)}$ are coefficients of cubic spline function which are varied for different α_k.

By solving the cubic spline function, the parameters A_i and B_i for different ellipsoids are calculated, which are shown in Table 1.

Table 1. Parameters used in the multiple ellipsoid model at a collision energy, $E_{\rm r}$ = 190 meV; V_i, V_i' and V_i'' correspond to assigned potential energies for 13, 8, 5 ellipsoid equipotential surfaces respectively.

A_i/Å	B_i/Å	V/meV	V_i/meV	V_i'/meV	V_i''/meV	i
6.20	5.42	1.0	1.9	2.9	5.9	1
5.92	5.19	2.0	2.9			2
5.76	5.03	3.0	3.5	5.7		3
5.65	4.91	4.0	4.2			4
5.56	4.82	5.0	5.0	6.0		5
5.49	4.74	6.0	6.0		9.9	6
5.42	4.67	7.0	7.0	8.0		7
5.30	4.55	9.0	11.0	14.2		8
5.24	4.50	10.0			30.0	9
5.02	4.31	15.0	15.0	29.0		10
4.86	4.16	20.0	27.0			11
4.45	3.78	40.0	45.0	54.0	50.0	12
4.14	3.45	70.0	98.0			13
3.98	3.23	100.0	190.0	190.0	190.0	14

It is acknowledged that IOS approximation is a more accurate theoretical method to calculate the collisions

between Ne atom and Na_2 molecule.[8] In this paper, the multiple ellipsoid model is applied to the ^{20}Ne–Na_2 system, and integral cross sections for the process $J = 0 \rightarrow J'$ at the incident energy of 190 meV are calculated. The results are given in Table 2.

Table 2. Integral cross sections $\sigma(J = 0 \rightarrow J')/\text{Å}^2$ of rotational excitation for ^{20}Ne–Na_2 system at the incident energy of 190 meV.

J'	IOS cross sections	cross sections of thirteen-ellipsoid model	cross sections of eight-ellipsoid model	cross sections of five-ellipsoid model
2	–	–	–	–
4	10.44	9.00	7.60	5.79
6	7.79	7.86	7.09	5.74
8	6.23	6.23	6.25	5.47
10	5.25	5.25	5.30	4.89
12	4.60	4.58	4.74	4.48
14	4.00	3.94	4.23	4.21
16	3.68	3.74	3.70	3.70
18	3.00	2.97	3.08	3.29
20	2.71	2.74	2.68	2.63
22	2.25	2.15	2.42	2.24
24	1.90	1.97	1.81	1.63
26	–	1.87	1.78	1.77
28	–	1.75	1.53	1.54
30	–	1.74	1.59	1.59
32	–	1.60	1.61	1.61
34	–	1.45	1.44	1.44
36	–	1.44	1.40	1.39
38	–	1.30	1.38	1.38
40	–	1.27	1.20	1.20
42	–	1.19	1.09	1.09
44	–	0.98	0.90	0.90
46	–	0.80	0.75	0.75
48	–	0.52	0.52	0.52
50	–	0.10	0.10	0.10

From Table 2, it can be seen that the excited cross sections can be reproduced with a model involving a small number of representative ellipsoids, although use of the multiple ellipsoid model is not sufficient to reproduce the $\sigma(J = 0 \rightarrow 4)$ cross sections, the thirteen-ellipsoid model gives very good results for the remaining cross sections. For example, when $J' = 6$, 8, 10, 12, the IOS integral cross section values of rotational excitation are 7.79, 6.23, 5.25, 4.60 Å^2 respectively, while the corresponding values of thirteen-ellipsoid model are 7.86, 6.23, 5.25, 4.58 Å^2 respectively. Moreover, the highest rotational excitation state $J_{\max}$ of our paper is 50, and the corresponding integral cross section values of rotational excitation are 0.10 Å^2. As shown in Table 2, use of the thirteen-ellipsoid model gives better results than the eight-ellipsoid model, and use of the eight-ellipsoid model gives better results than the five-ellipsoid model. In a word, the higher the number of ellipsoids used, the better results can be given. This is because when more ellipsoid equipotential surfaces are used, they can more accurately represent the impact of different regions of the intermolecular potential of Ne–Na_2 system for integral cross sections of rotational excitation. Moreover, because MC trajectories are 10^6, more realistic simulation of the actual situation of collisions between ^{20}Ne atom and Na_2 molecule can be given. Therefore, when the multiple hard-ellipsoid model is used to investigate the collision between Ne atom and Na_2 molecule, enough numbers of ellipsoid equipotential surfaces and MC trajectories should be chosen in order to obtain better integral cross sections of rotational excitation.

From the above calculation, we can see that the multiple hard-ellipsoid model is a good method to investigate the collision between Ne atom and Na_2 molecule. Therefore, the multiple hard-ellipsoid model is applied to the ^{16}Ne, ^{28}Ne, ^{34}Ne–Na_2 collision systems, and the integral cross sections of rotational excitation at the incident energy of 190 meV are calculated. The results are shown in Fig. 2.

According to Fig. 2, as the incident energy E_r is determined, the change rules of the state-to-state integral cross sections σ are quite similar: σ decreases gradually with the increasing rotational angular quantum number J'; the maximum integral cross section is $\sigma(J = 0 \to J' = 4)$; the form of the curve is negative exponential decay generally. It reveals that the rotational transfer probabilities of lower rotational excitation states are more than the higher rotational excitation states. Moreover, for the same rotational state J', the integral cross sections of lower rotational states decrease gradually with the increase of reduced mass of system, while the integral cross sections of higher states go up with the increase of reduced mass of system. For example, for $J' = 6$, it can be seen clearly from Fig. 2 that integral cross sections of ^{16}Ne, ^{20}Ne, ^{28}Ne, ^{34}Ne–Na_2 systems are 8.03, 7.86, 7.59, 7.47 Å^2, respectively; for $J' = 46$, these integral cross sections are 0.31, 0.80, 1.01, 1.13 Å^2, respectively.

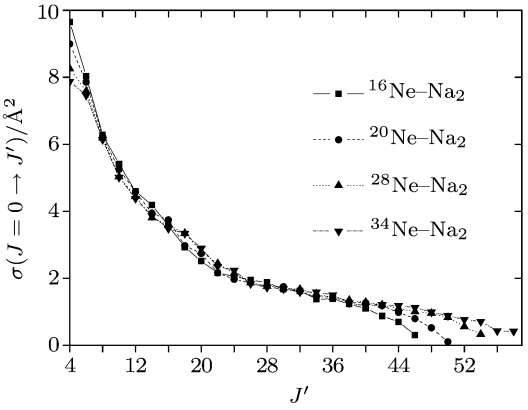


Fig. 2. Integral cross sections for rotational excitation in ^{16}Ne, ^{20}Ne, ^{28}Ne, ^{34}Ne–Na_2 collisions.

As relative translational energy $E_r = 190$ meV, total integral cross sections for rotational excitation in ^{16}Ne, ^{20}Ne, ^{28}Ne, ^{34}Ne–Na_2 collisions are given in Table 3.

As relative translational energy $E_r = 190$ meV, we also investigate how the multiple ellipsoid model can be used to investigate the contributions of different regions of the potential to inelastic cross sections for ^{20}Ne–Na_2 collision system. The results are given in Fig. 3.

Table 3. The total integral cross sections for rotational excitation in ^{16}Ne, ^{20}Ne, ^{28}Ne, ^{34}Ne–Na_2 collisions.

	^{16}Ne–Na_2	^{20}Ne–Na_2	^{28}Ne–Na_2	^{34}Ne–Na_2
σ_{tot}/Å^2	75.57	75.64	76.47	76.58

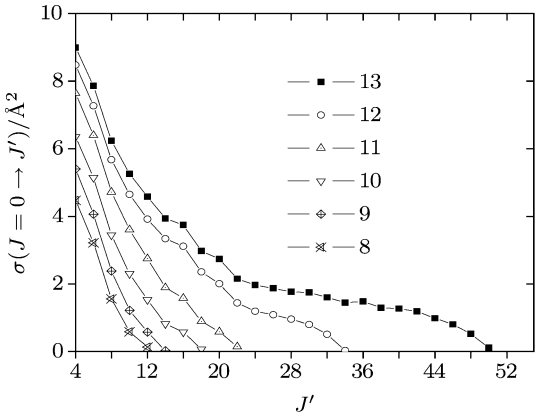


Fig. 3. Contributions made by different regions of the intermolecular potential to ^{20}Ne–Na_2 integral cross sections at $E_r = 190$ meV.

The cross section curves $i = 8, 9, 10, 11, 12, 13$ in the figure are obtained from the 0–7 meV, 0–11 meV, 0–15 meV, 0–27 meV, 0–45 meV, 0–98 meV and 0–190 meV regions of the intermolecular potential. In other words, the cross section curves $i = 8, 9, 10, 11, 12, 13$ correspond to the contributions of equipotential ellipsoid surfaces (1–8)-th, (1–9)-th, (1–10)-th, (1–11)-th, (1–12)-th, (1–13)-th to integral cross sections. According to Fig. 3, most of the contributions of low-ΔJ cross sections are from outer ellipsoids; most of the contributions of high-ΔJ cross sections are from inner ellipsoids. For example, scattering from the (1–8)-th ellipsoids contribute more than 50% to the total $\Delta J = 4$ cross section. However, the (1–12)-th ellipsoids contribute about 55.1% to the total $\Delta J = 24$ cross section, while the (1–10)-th ellipsoids contribute roughly none to it, in other words, the 11-th, 12-th ellipsoids contribute about 55.1% to the total $\Delta J = 24$ cross section. The higher the rotational excitation state is, the fewer the number of ellipsoids contributed to the corresponding cross section is. For example, the number of ellipsoids which contribute to $\sigma(J = 0 \to J' = 22)$, $\sigma(J = 0 \to J' = 32)$ and $\sigma(J = 0 \to J' = 45)$ is 3, 2, 1, respectively. However, it is important to note that contributions to the $\Delta J = 4$ cross section arise from scattering by a large range of the intermolecular potential (both high-energy and low-energy regions), whereas high ΔJ

transitions arise only from the inner, high-energy regions of the potential. It reveals that integral cross sections of rotational excitation are mainly affected by the regions of anisotropy of the repulsive intermolecular potential. The low-ΔJ cross section is a major subject for the influence of the outer ellipsoids or long-range repulsive regions of the potential. However, the high-ΔJ cross section is a major subject for the influence of the inner ellipsoids or short-range repulsive regions of the potential.

4. Conclusions

At the incident energy of 190 meV, the integral cross sections for rotational excitation of ^{16}Ne (^{20}Ne, ^{28}Ne, ^{34}Ne)–Na_2 collision systems by using multiple hard-ellipsoid model were calculated. Moreover, the change laws of the integral cross sections with the reduced mass of the system have been obtained. Finally, at this relative incidence energy, we investigated the influence of different regions of the intermolecular potential on integral cross sections of rotational excitation in ^{20}Ne–Na_2 collision. We can summarize the results as follows: as the relative incident energy of atom is determined, the integral cross sections of rotational excitation decrease gradually with the increase of rotational angular quantum number J', and the curve form is negative exponential decay generally. Moreover, for the same rotational state J', the integral cross sections of lower states decrease gradually with the increase of reduced mass of system, while the integral cross sections of higher states go up with the increase of reduced mass of system. It also reveals that integral cross sections of rotational excitation are mainly affected by the regions of anisotropy of the repulsive intermolecular potential. The low-ΔJ cross section is major subject for the influence of the outer ellipsoids or long-range repulsive regions of the potential. However, the high-ΔJ cross section is a major subject for the influence of the inner ellipsoids or short-range repulsive regions of the potential.

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