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Collision-induced interaction cross sections of 1–7 MeV B_2 ions incident on an N_2 gas target

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Abstract

Collision-induced interaction cross sections were measured for 1–7 MeV diatomic boron molecular ions incident on a nitrogen gas target by using a differentially pumped gas cell. The dissociation cross section of B_2^{q+} ($q = 1–3$) were measured to be on the order of 10^{-15} cm². The dissociation cross section was found to decrease with increasing ion charge state, and with increasing collision energy. The single-electron-loss cross section of B_2^{1+} was measured to be on the order of 10^{-16} cm², and the double-electron-loss cross section of B_2^{1+} was measured to be on the order of 10^{-17} cm² for 3 MeV ions. The single-electron-capture cross section of B_2^{2+} was measured to be on the order of 10^{-20} cm².

1. Introduction

The observation of triply charged diatomic boron molecules B_2^{3+} was reported in 1991 [1], representing the lowest mass diatomic molecular ion found in a 3+ charge state. The collision-induced interaction cross sections for this molecule are measured in the present work. When swift B_2 molecular ions pass through a gas target, they undergo various scattering processes such as dissociation, electron loss, and electron capture. Since the molecular ions are so weakly bonded (with bonding energies of a few eV), they dissociate when they collide with a target thicker than a few monolayers. Moreover, once multiply charged B_2^{q+} molecular ions are produced inside the gas target, they begin to dissociate with a characteristic lifetime. In the work reported here, B_2^{q+} molecular ion intensities were measured for each charge state for various nitrogen gas thicknesses using an Accelerator Mass Spectrometry (AMS) beamline on a tandem accelerator. Interaction cross sections were determined from these intensity measurements.

There are few published cross section measurements for molecular ions, in contrast with atomic ions. Although atomic ions can be fully stripped and remain stable, molecular ions are typically very unstable [2]. However, some diatomic molecular ions have been observed in charge states as high as 3+, for example, B_2^{3+} [1] and S_2^{3+} [3]. At present, there is no comprehensive theoretical model to calculate the molecular dissociation cross section at colli-

sion energies in the MeV regime and few other experimental works are reported. An exception is, for example, the work of Ben-Itzhak et al. [4] who measured the cross section for $NeAr^{2+}$ production from the charge stripping of a 900 keV $NeAr^{+}$ beam in an Ar gas target to be $(3 \pm 2) \times 10^{-18}$ cm².

2. Experimental methods

A schematic diagram of the AMS system used to make these measurements is shown in Fig. 1. Negative B_2^{-} ions produced in a sputter-type ion source [(4) in Fig. 1] were pre-accelerated to modest energies (69 keV), and then magnetically momentum per charge analyzed [Eqs. (6) and (8) in Fig. 1], before being injected into a tandem accelerator.

These negatively charged ions were stripped by an N_2 gas stripper to a positive charge state at the accelerator high voltage terminal, and the resulting high energy B_2^{q+} molecular ions emerging from the accelerator were directed through a differentially pumped gas cell located after the accelerator [(G) in Fig. 1]. A gas cell was used since a conventional foil stripper presents too thick a target, and the pressure (and hence gas thickness) could be monitored and controlled externally for the gas cell. The pressure in the gas cell was adjusted by a variable leak, and was monitored by a capacitance manometer. In each measurement, the pressure inside the gas cell was main-

Fig. 1. Schematic diagram of the AMS system at the University of North Texas: 1) CHIMERA; 2) target chamber; 3) Alphatross; 4) SNICS; 5) einzel lens; 6) 30° switching magnet; 7) beam slits/Faraday cup; 8) 90° analyzing magnet; 9) positive voltage terminal and N₂ gas stripper canal; 10) quadrupole lens; 11) 40° analyzing magnet; 12) 45° electrostatic analyzer; 13) retractable carbon foil; 14) solid state detector; and G) differentially pumped gas cell.

tained below 20 mTorr ($\sim 9 \times 10^{15}$ atoms/cm²) to ensure single collision conditions [5]. After passing through the gas cell, the ions were magnetically and electrostatically analyzed before being detected and energy analyzed in a solid state detector.

3. Dissociation model

Once highly charged metastable molecular ions are produced through a collision with a target inside the gas cell, they spontaneously dissociate, a process that is characterized by the lifetime. The probability that an incident ion will scatter in the gas cell with the target gas of number density n in an interval dx is

$$\mathcal{P}(x) dx = \sigma n dx, \quad (1)$$

where σ includes all the possible interaction processes that decrease the population of transmitted ions. Whenever incident particles scatter in the gas cell, their population (or intensity) decreases as

$$\frac{dI(z)}{dz} = -\frac{I(z)\mathcal{P}(z)}{n} = -\sigma I(z), \quad (2)$$

where z is an areal number density defined as nx . If, however, other processes are involved that increase the population of molecular ions with charge state q , e.g., either by electron capture or electron loss, the above differential equation must be modified to include those processes as follows:

$$\frac{dI^{(q)}(z)}{dz} = -\sigma^{(q)}I(z) + \sum_{q' \neq q} \sigma^{(q \leftarrow q')} I^{(q')}(z), \quad (3)$$

where $\sigma^{(q \leftarrow q')}$ indicates a cross section, either for electron capture or electron loss, for a change from a molecular charge state q' to a charge state q .

Since some of the molecular ions are in excited electronic states that are bound, they undergo a dissociation

process, i.e., natural dissociation with lifetime τ in analogy to radioactive decay. The surviving molecular ion intensity actually measured at the solid state particle detector located at the end of the AMS beamline is given by

$$I_{\text{det}}^{(q)} = f^{(q)} I^{(q)} e^{-t^{(q)}/\tau^{(q)}}, \quad (4)$$

where $I^{(q)}$ is the number of ions coming out of the gas cell with charge state q and $f^{(q)}$ denotes the geometric transmission factor through the beamline. The flight time of an ion with mass M and energy E can be calculated from

$$t^{(q)} = \frac{l_g}{v} = l_g \sqrt{\frac{M}{2E}}, \quad (5)$$

where l_g is the total distance that the ion travels to the detector after interacting inside the gas cell, and v is the molecular ion velocity.

When molecular ions with charge state q collide with gas molecules in the gas cell, the population in the transmitted beam will decrease as the pressure (or gas target thickness) in the gas cell increases, whereas the population of other charge states such as $q+1$ or $q-1$ will initially increase at lower pressure through electron loss or electron capture processes. But these other charge state populations will also decrease as the pressure goes up due to their own dissociation and decrease of charge state q . Therefore, for the measurement of the interaction cross section, the transmitted ions were measured instead of the scattered ions.

Fig. 2 shows the intensities of molecular charge states ($q=1-3$) as a function of nitrogen gas pressure in the gas cell, when 1.5 MeV $^{10}\text{B}^{11}\text{B}^{1+}$ ions were injected. Intensities were measured with a particle detector at the end of the AMS beamline. The intensities were normalized to -1 nA of injected B_2^- beam current going into the accelerator. Each measured intensity data set provides the cross section for a different process: the change in the $^{10}\text{B}^{11}\text{B}^{1+}$ intensity is due to the total interaction (dissociation, electron loss, and electron capture), the $^{10}\text{B}^{11}\text{B}^{2+}$ intensity is due to single electron loss, and the $^{10}\text{B}^{11}\text{B}^{3+}$ intensity is due to double electron loss.

Fig. 2. Intensities of each molecular charge states as a function of N₂ gas pressure when 1.5 MeV $^{10}\text{B}^{11}\text{B}^{1+}$ ions were injected.

For single collisions, two processes need to be considered: charge-conserving processes (total interaction) and charge-changing processes (electron loss or electron capture).

3.1. Charge-conserving ($AB^{q+} \rightarrow AB^{q+}$) process

For the charge conserving process (e.g., $B_2^{1+} \rightarrow B_2^{1+}$, $B_2^{2+} \rightarrow B_2^{2+}$, etc.), the rate equation for surviving molecular ions when molecular ions with charge state q are injected can be expressed by Eq. (2) with the cross section replaced by $\sigma^{(q)}$, which is the total interaction cross section due to all scattering processes including dissociation, electron loss, and electron capture. The solution of the above equation is

$$I^{(q)}(z) = I_0^{(q)} e^{-\sigma^{(q)}z}, \quad (6)$$

where $I_0^{(q)}$ is the number of molecular ions injected into the gas cell. Since the ions are metastable, they decay in analogy to the radioactive decay law. The actual number of ions detected at the particle detector is reduced as described by Eq. (4). The total interaction cross section $\sigma^{(q)}$ can be calculated by fitting the experimental data curve with the exponential functional form of Eq. (4).

3.2. Charge-changing ($AB^{q'+} \rightarrow AB^{q+}$) process

The rate equation for the charge-changing process, with incident charge state q' and exiting (measured) charge state q , can be written as

$$\frac{dI^{(q)}(z)}{dz} = -\sigma^{(q)}I^{(q)} + \sigma_{l,c}^{(q \leftarrow q')} I_0^{(q')} e^{-\sigma^{(q')}z}, \quad (7)$$

where the previously calculated Eq. (6) was used for $I^{(q')}$. The two subscripts l and c in $\sigma_{l,c}^{(q \leftarrow q')}$ indicate the *electron loss cross section* and the *electron capture cross section* from the molecular charge state q' to q , respectively. By substituting a trial solution that is a sum of exponential functions, the following solution is found:

$$I^{(q)}(z) = \frac{\sigma_{l,c}^{(q \leftarrow q')} I_0^{(q')}}{\sigma^{(q)} - \sigma^{(q')}} (e^{-\sigma^{(q')}z} - e^{-\sigma^{(q)}z}). \quad (8)$$

In the low pressure region, where $\sigma^{(q)}z \ll 1$ and $\sigma^{(q')}z \ll 1$, the above solution may be approximated to second order as

$$I^{(q)}(z) \approx \sigma_{l,c}^{(q \leftarrow q')} I_0^{(q')} \left[z - \frac{1}{2} (\sigma^{(q)} + \sigma^{(q')}) z^2 \right]. \quad (9)$$

The first few data points in the low pressure range of the measured intensity can thus be fitted quadratically.

By comparing the coefficients of the expansion and algebraically manipulating the equation one obtains the expression

$$\sigma_{l,c}^{(q \leftarrow q')} = \frac{f^{(q')}}{f^{(q)}} \frac{e^{-t^{(q')}/\tau^{(q')}}}{e^{-t^{(q)}/\tau^{(q)}}} \frac{A_{qq'}}{A_q} \frac{P}{z}, \quad (10)$$

where A_q and $A_{qq'}$ are the empirically determined factors for Eqs. (6) and (9), respectively. The geometric transmission factors $f^{(q)}$ and $f^{(q')}$ of charge state q and q' respectively were assumed to be equal for the calculation. Because the velocity is not changing in the charge changing process, the flight times $t^{(q)}$ and $t^{(q')}$ are the same and will cancel. The mean lifetimes of the 3 MeV boron diatomic molecular ions were measured to be $\tau(^{10}B_2^{1+}) = 200.1 \pm 8.5 \mu s$, $\tau(^{10}B_2^{11B^{2+}}) = 72.4 \pm 2.4 \mu s$, and $\tau(^{10}B_2^{3+}) = 4.5 \pm 0.1 \mu s$.

4. Results and discussion

The cross sections for the diatomic boron molecular ions interacting with a nitrogen gas target are summarized in Table 1.

The interaction cross sections were measured to be on the order of 10^{-15} cm^2 for all three charge states. The higher charge states appear to have slightly smaller interaction cross sections. The single-electron loss (or single ionization) cross section $\sigma_l^{(2 \leftarrow 1)}$ was measured to be on the order of 10^{-16} cm^2 , and the double-electron loss (or double ionization) cross section $\sigma_l^{(3 \leftarrow 1)}$ was measured to be on the order of 10^{-17} cm^2 . The single-electron capture cross section $\sigma_c^{(1 \leftarrow 2)}$ was measured to be on the order of 10^{-20} cm^2 , which implies a small branching ratio for this process. A few counts were observed for double-electron capture but not at a high enough intensity for cross section measurements. Since the interaction cross section is at least an order of magnitude larger than both electron loss and electron capture, the dominant process in the scattering is dissociation. Therefore, the dissociation cross sections can be approximated by the interaction cross sections.

Finally, the dissociation cross sections of the diatomic boron molecular ions, when colliding with a nitrogen target, were measured as a function of energy as shown in Fig. 3. The results show that the dissociation cross sections

Table 1

Cross sections of $^{10}B^{11}B$ ions in collision with an N_2 gas target at 3 MeV. The units are in cm^2

Energy	Injected	Transmitted		
		$^{10}B^{11}B^{1+}$	$^{10}B^{11}B^{2+}$	$^{10}B^{11}B^{3+}$
3 MeV	$^{10}B^{11}B^{1+}$	$(1.26 \pm 0.04) \times 10^{-15}$	$(2.50 \pm 0.87) \times 10^{-16}$	$(2.48 \pm 0.28) \times 10^{-17}$
	$^{10}B^{11}B^{2+}$	$(3.09 \pm 0.33) \times 10^{-20}$	$(1.15 \pm 0.02) \times 10^{-15}$	$(7.38 \pm 0.57) \times 10^{-19}$
	$^{10}B^{11}B^{3+}$		$(1.13 \pm 0.10) \times 10^{-17}$	$(1.07 \pm 0.02) \times 10^{-15}$

Fig. 3. The dissociation cross sections of diatomic boron molecular ions when colliding with a nitrogen gas target as a function of energy.

of the lower charge states have the higher value at a given collision energy, and the dissociation cross sections decrease as the collision energy increases.

In order to gain insight into the processes, the scale of the interaction distance may be considered. The scale of the interaction distance can be estimated from the cross section; thus, $a \sim \sqrt{\sigma/\pi}$. From the curve fit of the dissociation cross sections in Fig. 3, the approximate interaction distance is calculated to be $a \sim 2.0$ Å at 3 MeV. Therefore, the characteristic interaction distance is on the order of 1.3 times the B–B bond distance of approximately 1.6 Å calculated using GAUSSIAN [6]. This implies that a “good” theoretical description must include at least the three center problem (B–B–N). By using the Molière screened Coulomb potential [7], the interaction potential between B and N is calculated to be approximately 10 eV at a distance of 2.0 Å, which is sufficient to cause dissociation of the molecular ion. Therefore, an interaction distance corresponding to the measured cross section does suggest that the large value (on the order of a gigabarn) is plausible.

Single electron loss cross sections are on the order of 10^{-16} cm², which implies an interaction involving “non-bonding” core electrons (interaction distances $\leq 80\%$ of the bond distance). That the electron capture cross section is much smaller than the other cross section is not unexpected, since the electron in the target atom must be moving at a velocity comparable to the velocity of the ion in order to be captured as is rarely the case. At present, there is no comprehensive theoretical model to calculate the molecular interaction cross sections at these energies. It is hoped that the present observations will serve as stimuli to theorists to investigate the dissociation phenomena, and that a realistic theory of dissociation cross sections will result.

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References

- [1] D.L. Weathers, F.D. McDaniel, S. Matteson, J.L. Duggan, J.M. Anthony and M.A. Douglas, Nucl. Instr. and Meth. B 56/57 (1991) 889.
- [2] P.J. Bruna and J.S. Wright, J. Phys. B 26 (1993) 1819.
- [3] L. Morvay and I. Cornides, Int. J. Mass Spectrom. Ion Processes 62 (1984) 263.
- [4] I. Ben-Itzhak, I. Gertner and B. Rosner, Phys. Rev. A 47 (1993) 289.
- [5] F. Hopkins, R. Brenn, A.R. Whittemore, N. Cue, V. Dutkiewicz and R.P. Chaturvedi, Phys. Rev. A 13 (1976) 74.
- [6] M.J. Frisch et al., GAUSSIAN 92 (Gaussian, Inc., Pittsburg PA, 1992).
- [7] L.C. Feldman and J.W. Mayer, Fundamentals of Surface and Thin Film Analysis (North-Holland, New York, 1986).