Charge Transfer Cross Section for Gadolinium[†]

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Symmetric resonance charge transfer cross sections between singly ionized gadolinium ion and the parent atom were measured in the impact energy range of 100~1000 eV. The cross sections were determined from the ratio between the number of ions produced by charge transfer and those in primary ion beam. The primary ion beam of gadolinium was produced by a laser ion source in which gadolinium atoms were ionized by resonance photoionization. The slow ions produced by charge transfer and fast primary ions were detected with Faraday cups. The obtained cross section was about $1.5 \times 10^{-14} \text{ cm}^2$ at impact energy of 500 eV. The dependence of the measured cross sections on the impact energy roughly agrees with theoretical prediction.

1. Introduction

A charge transfer reaction is an important process in a many fields which use plasma processes, since the charge transfer process has a large cross section. A symmetric charge transfer process between ion and its parent atom is more important because the process has a large cross section even at a low energy. However, most experimental data for the symmetric charge transfer process are taken for rare gases^{1), 2)} and alkali metals.³⁾ There are few experimental data^{4), 5)} for atoms of transition elements. Recently, Sakabe *et al.*⁵⁾ have obtained the symmetric charge transfer cross section for gadolinium as a function of impact energy in the range of 10~1000 eV from the ratio of the charge transferred ions to the primary ions and the attenuation rate of the primary ions in the atomic beam. The cross sections obtained by them are larger than those of other elements studied in the past^{1)~3)} and are inversely proportional to the impact velocity below the impact energy of 300 eV.

In Atomic Vapor Laser Isotope Separation (AVLIS),⁶⁾ one specified isotope species is selectively photoionized by pulsed lasers and the produced ions are collected on electrodes. The charge transfer process between the produced ions and the atoms of another species degrades the enrichment factor. In AVLIS for the metallic elements such as gadolinium, efficient laser isotope separation requires a high density atomic vapor. However, the density is limited by degradation of the enrichment factor due to the process of the charge transfer. If the charge transfer cross sections obtained by Sakabe *et al.* are true, only low density atomic vapor is allowed in AVLIS for the metallic elements and efficient laser isotope separation is not feasible.

[†] Preliminary results of this work were reported at the 32nd Vacuum Symposium (Tokyo, 1991), but because of neglecting of a secondary electron emission due to bombardment of gadolinium ions, which was measured recently, the absolute magnitude of the cross sections presented at the symposium were 0~20% smaller depending on the impact energy than those present in this paper.

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Fig. 1. A schematic view of the experimental apparatus.

In the present work, the charge transfer cross sections for gadolinium atom were reinvestigated using a new cross-beam technique where a primary gadolinium ion beam was extracted from a laser ion source and slow ions produced by charge transfer collision were detected with a Faraday cup.

2. Experimental Apparatus

A schematic view of the experimental apparatus is shown in Fig. 1. The experimental apparatus consists of an atomic beam source region, a laser ion source region and a charge transfer collision region. The detailed diagrams of the laser ion source region and the collision region are shown in Fig. 2.

Gadolinium atomic beams were produced by heating gadolinium metal in the crucible with an electron beam. To eliminate the charged particles contained in the atomic beam, negative potential of 400 V was applied to elimination electrodes.⁷⁾ The atomic beams collimated by two slits entered the laser ion source and the collision region. The atomic density n in the collision region was estimated using the following equation

$$n = J_0 / \rho v \quad (1)$$

where J_0 is the deposition rate obtained from a quartz crystal sensor put at the height of 385 mm above the crucible, ρ is the density of bulk of gadolinium and v is the atomic beam velocity. The beam velocity obtained from the Doppler-shift of the laser fluorescence spectra was 600~900 m/s depending on the deposition rate.⁸⁾ Since the quartz crystal sensor was saturated in a short time because of a high deposition rate, another quartz crystal sensor was set at height of 973 mm above the crucible to know the atomic density in the collision region through the experiment over long time. The relation

Charge Transfer Cross Section for Gadolinium

Fig. 2. A detailed diagram of ion source region and collision region. (a) top view; (b) front view.

between the deposition rate at a height of 385 mm and that at a height of 973 mm is shown in Fig. 3. Recently, the deposition rate was measured with an electric microbalance.⁹⁾ The deposition rate agreed with that measured with the quartz crystal sensor. This result confirms that the deposition rate measured with the quartz crystal sensor is reliable. Depending on the input power of the electron beam, the atomic density in the collision region could be changed from $0.5 \times 10^{11} \text{ cm}^{-3}$ to $8 \times 10^{11} \text{ cm}^{-3}$.

In the laser ion source region, the atomic beam between electrode 1 and 2 was irradiated by a pulsed dye laser beam having wavelength of 370.94 nm. Gadolinium atoms were ionized by following one-wavelength two-photon ionization scheme.¹⁰⁾

The duration of pulsed laser was 10 nsec.

The ions produced by the pulsed laser beam constituted a plasma with released electrons. The plasma potential of this plasma was nearly equal to the higher potential

Fig. 3. Deposition rate and atomic density at a height of 385 mm as a function of the deposition rate at a height of 973 mm. Closed circles are the experimental values.

of those of the electrode 1 or 2.^{11), 12)} In this experiment, the potential of electrode 1, V_1 , was higher than that of electrode 2, V_2 . Thus, the ions extracted from the plasma impinged on the electrode 2 with the energy which corresponded to the difference between the potentials of electrodes 1 and 2, $e(V_1 - V_2)$. The electrode 2 had a hole of width 4 mm and height 8 mm. A small part of the ions was injected through the hole into the collision region.

The atomic beam in the collision region was collimated with the slit of width 25 mm and height 20 mm which was set at the height of 353 mm above the atomic beam source crucible. The profile of atomic beam in the collision region was estimated to be rectangular, 28 mm $\times$ 22 mm, from the shape of the slit and the height of the collision region from the crucible, considering that the atoms effused from an point source in the crucible. Since the width of the primary ion beam was narrower than that of atomic beam, all ions injected into the collision region passed in the atomic beam and were detected with Faraday cup 1. The surroundings of Faraday cup 1 and the electrode 3 were connected electrically and had the same potential V_2 with the electrode 2. Only the entrance of Faraday cup 2 was grounded. The entrance of Faraday cup 2 had the dimension of 55 mm $\times$ 60 mm and was much bigger than the dimension of the collision region. The potential distribution in the collision region was calculated using two-dimensional Laplace's equation with a personal computer. The potential distribution shown in Fig. 4 indicates that all slow ions produced by charge transfer reaction are expected to enter the entrance of the Faraday cup 2 and are detected by the Faraday cup 2. When too high potential V_2 was applied to the electrodes surrounding the collision region, the primary ion beam current detected by the Faraday cup 1 decreased because

of the deflection of the ion beam in a high electric field in the collision region. On the other hand, in the case of too low potential V_2 , the collision produced ion current can not be detected as an ion current pulse because it takes long time to collect the produced ions and the waveform of the ion current pulse is too small and too wide. The potential V_2 was determined so that the potential did not decrease the ion current detected with Faraday cup 1 and the produced slow ion current could be detected as a pulsed ion current with Faraday cup 2. It was also confirmed that the electric charge detected by the Faraday cup 2 did not depend on the potential in the neighborhood of the applied potential V_2 . The potential V_2 of $+30 \sim +100$ V was applied to the electrode 2 depending on the impact energy of $+100 \sim +1000$ eV.

Each Faraday cup consisted of a collection plate made of copper, the entrance of stainless steel mesh having transparency 80% and other five sides of stainless steel plates. The electric potential of -76 V was applied to the collection plates in order to collect all ions which enter the entrance grids. Recently we measured the yield of secondary electron emission from the collection plates due to bombardment of gadolinium ions. The secondary electron yield γ is represented approximately by the following equation,

$$\gamma = 3.8 \times 10^{-4} (E_i - 280) \quad \text{for } E_i > 280, \quad (2)$$

and

$$\gamma = 0 \quad \text{for } E_i \leq 280, \quad (3)$$

where E_i is the energy of gadolinium ions in unit of eV. This measurement will be described in detail.¹³⁾ The current detected by the Faraday cups include that due to the ions and the secondary electrons. In this experiment, the energy for the primary ion beam, E_i is equal to $V_1 + 76$ (eV), and that for the produced ions, E_i is equal to $0.8V_2 + 76$ (eV), where V_1 and V_2 are applied potential to the electrode 1 and the electrode 2, respectively. The factor 0.8 was estimated from the potential of the collision region shown in Fig. 4.

Fig. 4. The distribution of the potential in the interaction region. The potential distribution was calculated using two-dimensional Laplace's equation. Laplace's equation was numerically solved by Gauss-Seidel method with spatial grids of 23×30 . The curves are contour lines of constant potential plotted at every potential $0.1V_2$, where V_2 is the potential applied to the electrode 2.

Fig. 5. Typical oscillograms of the ion currents detected by the two Faraday cups. (a) Signal of primary beam ions and produced slow ions in the case of $V_1=400$ V and $V_2=100$ V. Atomic density at the collision region was $4.0 \times 10^{11} \text{ cm}^{-3}$. (b) Signal of primary beam ions and produced slow ions in the case of $V_1=150$ V and $V_2=50$ V. Atomic density at the collision region was $4.1 \times 10^{11} \text{ cm}^{-3}$.

3. Experimental Results

Typical oscillograms of the ion currents detected by the two Faraday cups are shown in Fig. 5. In the case of $V_1=400$ V, $V_2=100$ V and $V_1=150$ V, $V_2=50$ V, the ion currents are shown in Fig. 5(a) and (b), respectively. The electric charges obtained by integrating the ion current waveforms in Fig. 5 include the additional charge due to secondary electron emission. The electric charge of slow ions produced by charge transfer collision was obtained by dividing the electric charge, Q_{ex} , which was the integrated value of the ion current detected Faraday cup 2, by the factor $1+\gamma_{\text{ex}}$, where γ_{ex} is the yield of secondary electron emission from the collection plate in Faraday cup 2. The electric charge of the primary ion beam was obtained by dividing the electric charge, Q_i , which was the integrated value of the ion current detected with Faraday cup 1, by the factor $1+\gamma_i$, where γ_i is the yield of secondary electron emission from the collection plate in Faraday cup 1. Thus, the cross section σ is determined using following equation,

$$\sigma = \frac{Q_{\text{ex}}/(1+\gamma_{\text{ex}})}{Q_i/(1+\gamma_i)} \cdot \frac{1}{n \cdot l} = \frac{Q_{\text{ex}}}{Q_i} \cdot \frac{1}{n \cdot l} \cdot \frac{(1+\gamma_i)}{(1+\gamma_{\text{ex}})} \quad (4)$$

where n is the atomic density in the collision region and l is the interaction length. In Fig. 6, the ratio of Q_{ex} to Q_i is shown as a function of the atomic density in the case of $V_1=400$ V and $V_2=100$ V. The ratio of Q_{ex} to Q_i is proportional to the density in this experiment. The cross section is obtained from a slope of a line in Fig. 6, the interaction length l and the secondary electron emission yield, γ_i and γ_{ex} . In this case, the cross section is obtained to be $1.5 \times 10^{-14} \text{ cm}^2$. The ions in the primary ion beam are accelerated by a potential of about $0.2 V_2$ from the electrode 2 to the collision region, as shown in Fig. 4. Therefore, the impact energy of the primary ion beam, E_0 , is

$$E_0 = (V_1 - V_2) + 0.2 V_2,$$

in unit of eV. Fig. 7 shows the cross sections in the impact energy range of 100~

Fig. 6. A ratio of a charge detected by Faraday cup 2, Q_{ex} , to a charge detected by Faraday cup 1, Q_i , as a function of the atomic density in the collision region in the case of $V_1=400$ V and $V_2=100$ V.

Fig. 7. Cross section for symmetrical charge transfer on gadolinium as a function of the impact energy. Closed circles indicate the cross section obtained in our experiments. A solid curve and a thick dashed line indicate experimental data and the calculated results obtained by Sakabe *et al.* (ref. 5). A thin dashed line indicates the calculated results based on the theory by Rapp and Francis (ref. 16).

1000 eV.

4. Discussion

The obtained cross section data in Fig. 7 are scattered by 30~50% even at the same incident energy. The uncertainty of the measured data are considered to be due to following two origins. One origin is the electric charge of slow produced ions, Q_{ex} , which was obtained by integrating the slow ion current. The slow ion current was very small especially in the case of the impact energy lower than 200 eV and the background noise was not neglected as shown in Fig. 5(b). The uncertainty of the Q_{ex} was about 30% at the impact energy lower than 200 eV. Another origin is the atomic density in the collision region. The atomic density, n , is proportional to the ratio of deposition rate to beam velocity. Though accuracy of the beam velocity is considered to be $\pm 5\%$, the atomic beam density varied by $\pm 30\%$ within 1 sec.¹⁴⁾ However, the time constant of quartz crystal sensor was longer than 1 sec. Thus, the uncertainty of the atomic density, n , is considered to be about 30%. Therefore additional uncertainties of our cross section

measurement is 30% at the incident energy higher than 300 eV and 45% at the impact energy lower than 200 eV. The estimated errors are roughly equal to the spread of the cross section shown in Fig. 7.

In Fig. 7 are also shown the experimental and calculated results obtained by Sakabe *et al.*⁵⁾ They calculated the cross section of resonant charge transfer by impact parameter and close-coupling methods.¹⁵⁾ Their results are in fairly good agreement with the experimental results for the non-transition elements. Sakabe *et al.*⁵⁾ have suspected from the inconsistency between their experimental and calculation results for Gd that the charge transfer process for transition elements such as Gd is different from that of other elements. Calculation results based on the theory by Rapp and Francis¹⁶⁾ are also shown in Fig. 7. The cross sections measured by us are much lower than those by Sakabe *et al.*⁵⁾ and are not inversely proportional to the impact velocity even in the range of collision energy lower than 300 eV. Rather, our experimental results agree with the calculation results by Sakabe *et al.* and is about two times of those based on the theory of Rapp and Francis. It is considered that the charge transfer process for transition elements is also similar to that of other elements.

In AVLIS for transition metals such as gadolinium, we expect that a fairly high density atomic vapor is allowed and efficient laser isotope separation are not impossible in view point of degradation of the enrichment factor due to process of charge transfer reaction.

5. Conclusion

A resonance charge transfer between singly ionized gadolinium ion and the parent atom was investigated experimentally at an impact energy range from 100 eV to 1000 eV by detecting the slow ions produced by the charge transfer collision. The obtained cross section was about 1.5×10^{-14} cm² at an impact energy of 500 eV. The dependence of the cross sections on impact energy agrees roughly with the theoretical prediction.

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Charge Transfer Cross Section for Gadolinium

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Keywords

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