Population distribution subsequent to charge exchange of 29.85 keV Ni⁺ on sodium vapor

C. A. Ryder^a, K. Minamisono^{a,b,*}, H. B. Asberry^{a,c}, B. Isherwood^{a,b}, P. F. Mantica^{a,c}, A. Miller^{a,b}, D. M. Rossi^a, R. Strum^a

^a*National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, Michigan 48824, USA*

^b*Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824, USA*

^c*Department of Chemistry, Michigan State University, East Lansing, Michigan 48824, USA*

Abstract

The population of low-energy metastable states in Ni I subsequent to charge-exchange reactions and cascade decay was studied for 29.85 keV Ni ions impinging on a sodium vapor. The charge-exchange cross sections were calculated using a semi-classical time-dependent perturbation theory and redistribution of the population via rapid spontaneous decays was simulated. Collinear laser spectroscopy experiments were performed on four emission lines in Ni I to determine populations in low-energy metastable states: $3d^94s\ ^3D_3$ (204.787 cm⁻¹), $3d^94s\ ^3D_2$ (879.816 cm⁻¹), $3d^94s\ ^3D_1$ (1713.087 cm⁻¹) and $3d^94s\ ^1D_2$ (3409.937 cm⁻¹). Fair agreement was obtained between the simulation and experimental results. The low-energy metastable states were preferentially populated after the cascade decay in contrast to weak populations in near-resonant high-energy states in Ni I.

Keywords: Charge exchange, Nickel, Sodium vapor, Atomic population, Collinear laser spectroscopy

*corresponding author

Email address: minamiso@nscl.msu.edu (K. Minamisono)

1. Introduction

Collinear laser spectroscopy (CLS) [1] is a well-developed technique for measuring hyperfine (HF) spectra of ions or atoms. While laser excitation from the ground state of a singly ionized species using a commercial laser system may be possible for some elements, the electronic structure of the corresponding neutral species provides extended selection of transition frequencies for laser-spectroscopy studies. The conversion of an ion beam to an atomic beam via charge-exchange reactions with a suitable alkali-metal vapor [2, 3] is often used so that the atomic structure of the neutral species can be exploited for selection of its transition frequencies. The distribution of electronic states populated in the charge-exchange reactions is dependent on the first ionization energies of both the alkali-metal vapor and the state of interest in the incident atom. The distribution evolves with time due to rapid spontaneous decays, heretofore referred to as cascade decay, which redistribute the population towards lower energy levels. Low-energy metastable states not populated in the charge-exchange may be preferentially populated after the cascade decay. The population of low-energy metastable states may be greater than that of near resonantly populated states depending on the details of the cascade decay. These metastable states may be used as initial states for laser-excitation of atomic species where the ground-state excitation is not suitable for extraction of physics properties and/or the transition proves technically difficult. A larger population in the initial state leads to an improved signal-to-noise ratio, which is important in CLS measurements where only a limited number of atoms is available. One such example is the laser spectroscopy of radioactive isotopes, for which the production rate is limited.

Laser spectroscopy from metastable states populated in charge-exchange reactions has been exploited for noble gases [4], where the level density is sparse. In contrast, the level density of many other species, such as transition elements, is much higher. Depending on the level density, atomic lifetimes, and branching ratios of each transition within the cascade decay, the population of the outgoing atomic beam may (i) be fragmented across many levels, (ii) remain in a near-resonant metastable state, which is directly populated in the charge-exchange reactions, (iii) or be accumulated in a low-energy metastable state after the cascade decay. The near-resonant process has been previously studied using a Mn beam [5]. In this work, the population of low-energy metastable states was derived from laser spectroscopy on

a Ni atom beam. A more complete theoretical treatment for the cross sections of charge-exchange reactions is employed [6], and the time-dependent change of the population distribution in Ni I is simulated and compared to the population distribution probed by laser spectroscopy. The framework of the present study aids in future planning of spectroscopy on low-counting rate measurements, in particular on radioactive isotopes.

2. Atomic charge exchange

2.1. Asymmetric charge-exchange process

The overall charge-exchange process considered in the work is

where $\text{Ni}^+(^2D)$ is the incident ion beam in the electronic energy state 2D , $\text{Na}(^2S)$ is the alkali in its ground state 2S , $\text{Ni}(i)$ is the outgoing atom beam in the state i , $\text{Na}^+(^1S)$ is the alkali ion in its ground state 1S and ΔE is the energy difference between the final and initial electronic states. Alternatively, the entry energy of the Na electron relative to the Ni I ground state, as shown in Fig. 1, is defined as the difference of ionization potentials between $\text{Na}(^2S_{1/2})$ and $\text{Ni}(^3F_4)$; that is, $I(\text{Na}) - I(\text{Ni}) = 20115.5 \text{ cm}^{-1}$, where I is the ionization energy for elements in the parentheses. If the energy level of the $\text{Ni}(i)$ state happens to be at the entry energy, the charge-exchange reaction has $\Delta E = 0$ and is resonant. However, such a state does not exist in Ni I and the nearest state to the entry energy is the $3d^8 4s^2 \ ^1G_4$ state at $22102.349 \text{ cm}^{-1}$. When $\Delta E \neq 0$, the charge-exchange process is non-resonant and leads to an energy exchange between the initial and final states [3]. The non-resonant process in the asymmetric charge-exchange reaction considered here is more common than the resonant process, since the entry energy, defined by the choice of alkali species, cannot be varied freely.

2.2. Charge-exchange cross section

The cross section of asymmetric charge-exchange reactions was calculated using the semi-classical time-dependent perturbation theory developed by Rapp and Francis [6] with an additional correction by a factor of 2 within the argument of the sine function [7]. The cross section for charge transfer in a collision of an incident atom with velocity v and impact parameter b is given by

$$\sigma(v) = 2\pi \int_0^\infty P_\omega(b, v) b db \quad (2)$$

$$P_\omega(b, v) = f P_0(b, v) \text{sech}^2 \left[\frac{\omega}{v} \left(\frac{a_0 \pi b}{2\gamma} \right)^{1/2} \right] \quad (3)$$

$$P_0(b, v) = \sin^2 \left[\left(\frac{8\pi}{\gamma a_0} \right)^{1/2} \frac{2\pi I b^{3/2}}{h v} \left(1 + \frac{a_0}{\gamma b} \right) \exp \left(-\frac{\gamma b}{a_0} \right) \right] \quad (4)$$

where f is the statistical weight factor, I is the average of the ionization potentials of both Ni(i) and Na I, $\gamma = (I/13.6)^{1/2}$, a_0 is the Bohr radius and w is the difference of ionization potentials between the ground state Na and the state of interest in Ni. The cross section is expressed in terms of energy difference of the atomic level of interest in Ni from the entry energy of electron and the average ionization potential. The charge-exchange cross sections were calculated in this model for the neutralization of a Ni⁺ beam at an energy of 29.85 keV impinging on sodium atoms, as shown in Eq. (1). In the calculation performed here, the Ni ion, Na atom and Na ion are all assumed to be in their ground states, with the atomic configuration indicated in Eq. (1). The outgoing Ni(i) may reside in any one of the energy levels within Ni I depicted in Fig. 1. For each i state, a cross-section was calculated with and without the statistical weight factor (f). The results are shown in Fig. 2. The bare cross section, not including f , peaks at the entry energy of 20115.5 cm⁻¹. The fraction of collisions that produce the final state in Eq. (1), which represents charge transfer, is denoted by f and consists of contributions from the spin (f_S) and angular momentum (f_L) degeneracies for the {Ni⁺(²D) + Na(²S)} and {Ni(i) + Na⁺(¹S)} systems. The state nearest the peak in the bare cross section, the 3d⁸4s² ¹G₄ state in Ni I at 22102.349 cm⁻¹, does not have the largest cross section value because of a small $f = 0.25$ ($f_S \times f_L = 0.25 \times 1$).

3. Experiment

The CLS experiment was performed at the BEam COoling and LAsEr spectroscopy (BECOLA) facility [8] at the National Superconducting Cyclotron Laboratory at Michigan State University. The schematic of the experimental setup is shown in Fig. 3. Ion beams of Ni were generated using

a Penning Ionization Gauge (PIG) ion source [9]. The extracted Ni-ion beam was injected into a radio-frequency quadrupole (RFQ) cooler [10], where the Ni ions were cooled (improving beam emittance) through collisions with a helium-buffer gas, and extracted at an energy of 29.8558 ± 0.0020 keV. The cooled Ni^+ beam was transported to the BECOLA CLS beam line and laser light was collinearly overlapped with the Ni beam. A vertical charge-exchange cell (CEC) [3] containing sodium vapor was used to neutralize the Ni beam. The CEC reservoir temperature was maintained at about 700 K to achieve an overall neutralization efficiency of 50% and minimize broadening of the laser resonance line width due to inelastic charge-exchange reactions. A variable voltage was applied to the CEC to tune the velocity of the incoming Ni-ion beam into resonance with the Doppler-shifted laser frequency of the atomic transition of interest. Laser-induced fluorescence was collected using an ellipsoidal reflector [5] and detected by a photomultiplier tube, which was placed 40 cm downstream from the exit of the CEC. The fluorescence was detected as a function of the voltage applied to the CEC.

The PIG ion source is a new feature of the BECOLA facility and was essential for the production of the Ni ions beams used in this work. Details of the construction and operation of the PIG source are discussed in the next section.

3.1. Ni-ion beam production

A PIG ion source [9] was used to produce Ni-ion beams. A schematic layout of the ion source is shown in Fig. 4. The PIG source is a compact and efficient ion source used to produce ions of metallic elements, which have high melting points and low vapor pressures. The source consisted of a pair of Ni cathode and anticathode disks equidistant on either side of a central anode disk. An axially symmetric magnetic field generated by an electrostatic magnet was applied along the central axis of the anode and cathodes encompassing the plasma discharge region. The axial magnet produced a magnetic field strength of 500 G at 2.0 A (2.7 V). A potential difference of 600 V was applied between the cathodes and anode. An argon-buffer gas was used in the formation and perpetuation of the plasma. The pressure of the argon-buffer gas in the plasma region could not be directly measured due to the geometry of the source and was estimated to be about 10^{-3} Torr based on the pressure measured 0.5 m away from the PIG source. The ionized argon-buffer gas was accelerated through the anode hole and sputtered material from the two cathodes, which were both composed of nickel (purity >

99.95%). The sputtered nickel was then ionized by the fast-electrons within the plasma and extracted from the source through a concentric bored hole in the cathode (0.08 mm in diameter) by applying a -2.5 kV potential to a conically shaped extraction electrode. A large fraction of the extracted ion beam from the PIG source was composed of argon ions from the argon-buffer gas, which were removed by the RFQ cooler that effectively operates as a quadrupole mass filter [11] and has a mass resolving power of ~ 3 . The overall output ion-beam current was about $1\text{ }\mu\text{A}$ with approximately 1 nA of Ni-ion beam obtained after the RFQ cooler. The purity of the Ni beam after the RFQ cooler was estimated to be $>90\%$ based on fluorescence yield in laser spectroscopy measurements.

3.2. Laser system

Laser spectroscopy was performed on ^{58}Ni , which has the largest natural abundance of 68.0% of the stable Ni isotopes. Four transitions were chosen for spectroscopy because each of the four states (i) is a metastable state, which can be used as a long-lived lower level for spectroscopy, (ii) has a corresponding upper state, which decays directly back to the same lower level with a large branching ratio, (iii) has corresponding upper states with relatively strong Einstein coefficients, and (iv) had accessible laser frequencies using the BECOLA laser system. Details of the atomic transitions are listed in Table 1. The laser light for the Ni I spectroscopy was obtained from a Spectra Physics frequency doubler, which produced second-harmonic light. The fundamental light was obtained using a Spectra Physics single-mode Ti:Sapphire ring laser pumped by a Spectra Physics, continuous-wave 15 W, 532 nm laser. The laser light was transported to the CLS beam line through a polarization maintaining single-mode optical fiber and transmitted along the beam line collinearly with the Ni-ion beam. The laser power was set and maintained at $100\text{ }\mu\text{W}$ and stabilized with a $\pm 0.05\%$ power fluctuation using a laser-power controller [12]. The diameter of laser light at the fluorescence detector was $1.7 \pm 0.2\text{ mm}$ measured by a CCD camera.

4. Results

4.1. Simulation on initial population

The population distribution of Ni atoms at the fluorescence detector was simulated by taking into account the Ni-atom beam energy (29.85 keV), distance of the fluorescence detector from the CEC (40 cm), laser power (100

μW over 1.7 mm diameter), stimulated absorption and spontaneous transition probabilities, and branching ratios for each of the transition steps in the cascade decay. A relative distribution of the initial population immediately following the charge-exchange reactions was derived from the calculated cross sections using the model of Rapp and Francis [6] discussed in section II. The duration of the cascade decay was $1.26 \mu\text{s}$, which was the time the Ni-atom beam traveled between the CEC and the fluorescence detector. The ^{58}Ni -atom beam had a kinetic energy of 28.95 keV and a velocity of 3.152×10^7 cm/s. Energy levels, branching ratios and Einstein coefficients were taken from the Kurucz database [13] and used in the simulation. The derived population distribution is shown in Fig. 5, where the initial (pre-cascade) and final (post-cascade) populations are shown in open squares and solid circles, respectively. The total population among all energy levels was normalized to one.

The population within the states residing near the entry energy (20115.5 cm^{-1}) in the charge-exchange reactions remained the same or increased slightly after the cascade decay, since these states do not decay within the flight time and have minimal feeding from higher-energy states. The initial/final populations of these near-resonant metastable states in Ni I are at most 5% of the total population due to the high level-density (fractionalization of population) and the statistical weight factor. On the other hand, many states around 30000 cm^{-1} populated in charge-exchange reactions decay into low-energy metastable states, which are not initially populated. Whereas most low-energy metastable states initially have small populations ($< 0.2\%$), due to their large energy separation from the entry energy, some of low-energy metastable states, e.g., the states at 204.787 cm^{-1} and 879.816 cm^{-1} , accumulate 13% of the total population after the cascade decay. Numerical values of the normalized populations for the four low-energy metastable states are summarized in Table 2. Level information from the NIST atomic database [14] was used (summarized in Table 1) to check possible variations of the final population distribution from the Kurucz database and the deviation was taken into account as the systematic error.

4.2. Collinear laser spectroscopy on ^{58}Ni

A typical result of a CLS measurement for the $4s \ ^3D_3$ (204.787 cm^{-1}) $\leftrightarrow$ $4p \ ^3P_2$ ($28569.203 \text{ cm}^{-1}$) transition is shown by the solid circles in Fig. 6. The error on the data points represents the statistical error. The peak was

fitted using an asymmetric Voigt profile [15] to take into account asymmetric broadening of the resonance line shape due to inelastic charge-exchange processes [3], and the result is shown as a solid line in the figure.

The background-subtracted peak area was determined for each of the four Ni I emission lines. The peak area in each instance was normalized by the Ni-atom beam intensity and the duration of the measurement. Two additional corrections were applied to the normalized peak area. The first correction accounted for the photon emission probability at the fluorescence detector to allow for a comparison amongst the four transitions. The effective detection length was 2.5 cm, and the probability that one Ni atom passing through the detection region emits one photon within this length is less than one for the given velocity of the Ni-atom beam. An exponential decay with a decay constant given by the sum of the inverses of the Einstein coefficient and absorption probability, accounted for the difference in the emission probability as:

$$f_{\text{decay}} = \left[1 - \frac{N_i}{N_f}\right] = 1 - \exp \left[\frac{l_{\text{det}}}{\nu} \left(\frac{1}{A_{ij}} + \frac{g_j}{g_i P_{ij}} \right)^{-1} \right], \quad (5)$$

where N_f (N_i) represents the final (initial) population of the level, l_{det} is the length of detection region, A_{ji} is the Einstein coefficient for spontaneous decay, g_j (g_i) is the degeneracy of the upper (lower) state, and P_{ij} is the absorption probability given by

$$P_{ij} = \frac{2c^2}{\pi^2 \omega^3} \frac{I_L}{d^2} \tau \frac{A_{ij}}{h}. \quad (6)$$

Here, c is the speed of light, $\omega = 2\pi\nu$ represents the frequency of the transition, I_L is the laser power, d is the diameter of the laser beam, τ is the lifetime of the upper state, and h is Planck's constant. The second correction was for the losses due to laser-stimulated absorption and subsequent decay branches that lead to dark states during the flight time of Ni-atom beams. The simulation took spontaneous decays into account, but did not include laser stimulated absorption. The cycle of laser-stimulated absorption and spontaneous decay started once the Ni-atom beam was produced in the CEC and continued through the flight path between the CEC and the fluorescence detector. The number of the cycles, n , was determined using the decay constant as the time for one cycle. Decay losses were accounted for through the branching ratio β :

$$f_{\text{cyclic}} = \beta^{-n} = \beta^{-\frac{l_{\text{flight}}}{v} / \left(\frac{1}{A_{ij}} + \frac{g_j}{g_i P_{ij}} \right)} \quad (7)$$

In Equation 6, l_{flight} represents the distance between the CEC and the fluorescence detector.

5. Discussion

The deduced populations from the CLS measurements are shown together with the results of the simulation in Fig. 7. The experimental and simulated results for each energy level were normalized to the value for the 204.787 cm^{-1} state. The uncertainty on the experimental values were derived from the statistical error of the fit and the standard deviation of the baseline fluctuation of the fluorescence spectrum. The uncertainty of the simulated values were deduced from the deviation in the final population simulated with line information from the Kurucz database and those from the NIST database. The overall agreement of the relative populations in 204.787 cm^{-1} , 879.816 cm^{-1} and 3409.937 cm^{-1} states with the simulated values is excellent, given the simple framework of the cross-section calculation. No laser-induced fluorescence signal was detected for the $3d^9 4s \ ^3D_1$ (1713.087 cm^{-1}) $\leftrightarrow$ $3d^9 4p \ ^3P_0$ ($30192.251 \text{ cm}^{-1}$) transition, and an upper limit estimated from the background fluctuation is presented in the Fig. 7. A signal of the expected 3D_1 to 3P_0 transition was searched within $\pm 4.5 \text{ GHz}$ of the expected resonance frequency based on the Kurucz [13] and NIST [14] databases. The simulation predicts a larger population for the 1713.087 cm^{-1} state than that for the 3409.937 cm^{-1} state, which was observed in this work.

Contributions to the charge-exchange cross sections from excited states in either the Ni-ion beam or sodium vapor are not considered in the present simulation. The Ni-ion beam from the PIG ion source was cooled in the RFQ cooler, and metastable states were likely quenched. If the second-excited fine-state in Ni II ($3d^8 4s \ ^4F$) survived the cooling, the cross sections for charge exchange would be small due to the statistical weight factor for the spin configuration. Sodium excited states may be populated due to the high temperature of and Na-Na collisions in the sodium vapor. However, given the temperature of CEC reservoir of $\sim 700 \text{ K}$ and assuming a Maxwell-Boltzmann distribution of the thermal population, the contribution from the sodium excited is negligible.

The discrepancy between simulated and experimental values for the final population of the 1713.087 cm^{-1} state is unclear. However, this discrepancy should not take away from the good agreement obtained between simulation and experiment for populations in the other three low-energy metastable

states in Ni I. The present framework for the charge-exchange cross section calculation followed by cascade decay has certain predictability and can be used to plan future laser spectroscopy experiments. The preferentially populated low-energy metastable states after the cascade decay may be used as a lower level in laser spectroscopy, when the ground state transition is not suitable for the extraction of physics properties and/or the transition proves technically difficult. Also, the increased population of the low-energy metastable states compared to those of weakly populated states near the entry energy has a significant impact on measurement reliability, as well as data accumulation time in low counting rate radioactive isotope experiments.

The present framework of the simulation can be applied to other charge-exchange systems. Spectroscopic information on atomic energy levels and especially Einstein coefficients are critical for reliable simulations. Einstein coefficients are often missing for many transitions and for some elements, for example in Se, Tc, Br and I, little information is available. For radioactive isotopes of such elements, the population distribution simulations are critical for planning of new measurements on nuclear moments and charge radii, which are currently unknown. Experimental and theoretical works are highly encouraged on studies regarding atomic energy levels and Einstein coefficients.

6. Summary

The distribution of population among energy levels, produced via the charge-exchange reactions, was simulated for a 29.85 keV Ni-ion beam impinging on a sodium vapor. The cross sections of the charge-exchange reactions between Ni ion and sodium vapor in their ground states were calculated by a semi-classical perturbation theory. The subsequent population of low-energy electronic states in neutral Ni was simulated following cascade decay. The simulated populations were compared with populations in four low-energy metastable states in Ni I, $3d^9 4s\ ^3D_3$ (204.787 cm⁻¹), $3d^9 4s\ ^3D_2$ (879.816 cm⁻¹), $3d^9 4s\ ^3D_1$ (1713.087 cm⁻¹) and $3d^9 4s\ ^1D_2$ (3409.937 cm⁻¹), which were determined by laser spectroscopy measurements. The experimentally-deduced populations for three low-energy metastable states in Ni I were in good agreement with the simulation. The distribution of electronic states populated in the charge-exchange reactions is of interest because some of the metastable states may be used for laser spectroscopy, when the ground state transition is not suitable for the extraction of physics

properties and/or the transition proves technically difficult. Also, knowledge of sufficiently populated levels has a significant impact on low counting rate radioactive isotope experiments, in terms of measurement reliability and data accumulation time. The possibility of using the preferentially populated low-energy metastable states may lead to determinations of physics properties by utilizing new transition opportunities.

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Table 1: Atomic transitions in ^{58}Ni I used in the present study. The branching ratio (β_{ji}) and atomic lifetime (τ_j) were deduced from the NIST line information [14], and the Einstein coefficient A_{ji} from the Kurucz data base [13].

E_i (cm^{-1})	Term i	E_j (cm^{-1})	Term j	A_{ji}	β_{ji} (%)	τ_j (ns)
204.787	$3d^9 4s \ ^3D_3$	28569.203	$3d^9 4p \ ^3P_2$	1.0	91	9
879.816	$3d^9 4s \ ^3D_2$	29320.762	$3d^9 4p \ ^3F_3$	0.4	58	14
1713.087	$3d^9 4s \ ^3D_1$	30192.251	$3d^9 4p \ ^3P_0$	1.2	99	9
3409.937	$3d^9 4s \ ^1D_2$	31441.635	$3d^9 4p \ ^1D_2$	0.6	89	16

Table 2: The population (P) for each of the four low-energy metastable states in Ni I. The first column is the low-energy metastable states and the second, third and fourth columns are the initial and final populations, respectively.

E_i (cm^{-1})	P_{initial} (% of total)	P_{final} (% of total)	P_{final} (% of total) NIST
204.787	0.1	13.9	13.6
879.816	0.2	13.3	11.9
1713.087	0.2	8.7	8.7
3409.937	0.1	3.8	3.7

Figure 1: Energy levels of Ni I and Na I (partial) in terms of Ni I ground state. The solid red line is the ground state of Na I and the energy difference between ground states of Ni I and Na I is defined as the entry energy. The dashed red line is the entry point of the electron within Ni I.

Figure 2: Charge-exchange cross section for levels in Ni I. The solid line displays the bare charge-exchange cross section without f , while the solid circles show the cross section distribution with f for the discrete states in Ni I.

Figure 3: Schematic layout of the CLS experimental setup.

Figure 4: Cross section of the Penning ionization gauge ion source. The PIG source has a rotational symmetry around the central horizontal axis. The voltage applied to the cathode/anti-cathode was -450 V , while the anode was biased at $+150\text{ V}$.

Figure 5: Simulated normalized populations of the energy levels in Ni I are shown both pre-cascade (blue squares) and post-cascade (red circles). The low-lying metastable states are separately shown in the insert.

Figure 6: A typical spectrum of ^{58}Ni I for the $28,569.203\text{ cm}^{-1}$ to 204.787 cm^{-1} transition. The spectrum was fitted using an asymmetric Voigt profile. The error bars on the experimental values represent statistical errors.

Figure 7: Experimental and simulated relative populations of the four low-energy metastable states in Ni I. The uncertainty on the experimental values is from the statistical error and the standard deviation of the baseline fluctuation of the fluorescence spectrum. The uncertainty of the simulated values is from the deviation in the final population simulated with line information from the Kurucz database and those from the NIST database.