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B. G. Birdsey
University of Nebraska - Lincoln

H. M. Al-khateeb
Jordan University of Science and Technology

Timothy J. Gay
University of Nebraska - Lincoln, tgay1@unl.edu

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B. G. Birdsey*, H. M. Al-khateeb†, and T. J . Gay*

*Behlen Laboratory of Physics, University of Nebraska, Lincoln, NE 68588-0111 USA
†Department of Physics, Jordan University of Science and Technology, Irbid, P.O. Box 3030, 22110 Jordan

Abstract. We report on measurements of the integrated Stokes parameters of the light emitted from four well-LS coupled states of the $3p^4(^1D)4p$ manifold of ArII, following the simultaneous ionization and excitation of neutral argon by polarized electrons. As for all states, the state multipoles of J can be expanded in terms of the total orbital (L) and the total spin (S) state multipoles. By splitting each L and S state multipole into multipoles for the core and outer electron, we have experimentally obtained for the first time the normalized integrated state multipole of rank 4 (hexadecapole moment) for the $3p^4(^1D)$ core of ArII. We will comment on the Rubin-Bederson hypothesis as it pertains to this collision system [G. Csanak *et al.*, Comments At. Mol. Phys. **30**, 165 (1994)] as well as elucidate the data analysis techniques used.

INTRODUCTION

The theory of electron-atom interactions has been known in principle since the development of the Dirac equation, yet it is the tension between "what can be measured" and "what can be calculated" that has driven much of atomic collision physics to this day. Recently the convergent close coupling (CCC) technique of Bray and Stelbovics [1] has proven its efficacy in calculating electron interactions with simple atoms such as H [2], He and Na [3], and Li [4]. However the situation for the heavy noble gases is much more complicated; agreement between theory and experiment in this area ranges from adequate to dismal [5 - 8].

Our experiment is centered around the three body process of simultaneous ionization and excitation of Ar II. The specific process we examined is given by the reaction

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$$\begin{aligned} e^- + Ar\left(3p^6\right) \quad &\rightarrow Ar^{+*}\left(3p^4\left(^1D\right)4p\right) \quad + 2\ e^- \\ &\searrow Ar^{+*}\left(3p^4 4s \text{ or } 3p^4 3d\right) + \ \gamma \end{aligned} \quad , \qquad (1)$$

where the excited states of interest in the $3p^4(^1D)4p$ manifold are $^2F_{7/2}$, $^2F_{5/2}$, $^2D_{5/2}$, and $^2P_{3/2}$. The incident electron beam is transversely polarized and the scattered electrons are not detected. These particular residual ionic states are advantageous because they are known to be well-LS coupled [9], unlike neutral Ar. This simplifies the interpretation of our data and reduces the difficulty for theory to calculate electron-atom scattering processes [7, 8].

Our goal is to characterize the final state atomic charge cloud in terms of individual contributions from the $3p^4(^1D)$ ionic core and the excited 4p outer electron. The shape of the charge cloud is completely described by the tensor multipoles of the excited ion's density matrix [10]. As detailed in our earlier work [11], the tensor multipoles of L can be expanded as

$$\begin{aligned} \left\langle \mathcal{T}^{\dagger}_{KQ}(L)\right\rangle = (2L+1)\sum_{\substack{k_c q_c \\ k_o q_o}} \sqrt{(2k_c+1)(2k_o+1)} \begin{Bmatrix} l_c & l_o & L \\ l_c & l_o & L \\ k_c & k_o & K \end{Bmatrix} \times \\ \left(k_c q_c, k_o q_o \middle| KQ\right)\left\langle \mathcal{T}^{\dagger}_{k_c q_c}(l_c) \otimes \mathcal{T}^{\dagger}_{k_o q_o}(l_o)\right\rangle, \end{aligned} \qquad (2)$$

where the subscripts c and o refer to the multipoles associated with the core and outer electron. Notice that the multipole moments of the $\mathcal{T}_{k_c q_c}(l_c)$ and $\mathcal{T}_{k_o q_o}(l_o)$ sub-shells are generally not factorable, i.e. $\left\langle \mathcal{T}^{\dagger}_{k_c q_c}(l_c) \otimes \mathcal{T}^{\dagger}_{k_o q_o}(l_o)\right\rangle$ is not generally equal to $\left\langle \mathcal{T}^{\dagger}_{k_c q_c}(l_c)\right\rangle\left\langle \mathcal{T}^{\dagger}_{k_o q_o}(l_o)\right\rangle$. Therefore it is crucial to choose an experimental system where the multipole moments can be factored or where one can make enough observations to completely determine the coupled multipole moments. We introduce the following notation for the reduced multipole moments :

$$\begin{aligned} \ell(J,j)_{KQ,kq} &= \left\langle \mathcal{T}^{\dagger}_{KQ}(J) \otimes \mathcal{T}^{\dagger}_{kq}(j)\right\rangle \Big/ \left\langle \mathcal{T}^{\dagger}_{00}(J) \otimes \mathcal{T}^{\dagger}_{00}(j)\right\rangle \\ \text{and } \ell(J)_{KQ} \quad &= \left\langle \mathcal{T}^{\dagger}_{KQ}(J)\right\rangle \Big/ \left\langle \mathcal{T}^{\dagger}_{00}(J)\right\rangle, \end{aligned}$$

which will be used throughout the remainder of this work.

Because of our collision geometry, symmetry allows only a few of the multipoles of J to be non-zero [11]. Since these ionic states are well-LS coupled, L and S are good quantum numbers and the expansion of the multipoles of J into products of L multipoles and S multipoles is allowed [12]. We now invoke the Rubin-Bederson (RB) hypothesis [11], which states that if the collision time is significantly shorter than it takes for the excited system to "relax into its energy eigenstates", the collision can be considered as impulsively preparing each subsystem of the ion (the core, outer electron, and continuum electrons).

To determine whether the RB hypothesis should hold true it is necessary to check the appropriate time scales. The duration of the near threshold ionization/excitation process can be estimated by the time it would take an electron with the asymptotic energy of 2 eV to traverse three diameters of the residual ion, or ~$6x10^{-16}$s. This is a factor of 3 shorter than the "Coulomb relaxation time", the time necessary to couple l_c and l_o into the total L, conservatively gauged by the largest splitting in the $3p^4(^1D)\ ^2L$ manifold. The next time scale is the "fine structure relaxation time" which can be estimated from the energy splitting of the $^2F_{7/2}$ and $^2F_{5/2}$ fine-structure levels, and corresponds to 10^{-13}s. Therefore we would expect the RB hypothesis to hold in this experiment, if only marginally for decoupling L into l_c and l_o.

In our experiment, as well as the general case of atomic collisions, the fine-structure relaxation time is much longer than the collision time. According to the RB hypothesis, this implies that the subsystems of L and S can be described independently of each other, i.e. that the multipoles $t_{KQ}(L)$, $t_{KQ}(S)$ are properties of the manifold and are not properties of the individual states of the manifold. Assuming that this relationship is not then perturbed by terms that would convert angular momentum from one form to another (i.e. L to S), these multipoles of the manifold can be determined by observing a subset of the J multipoles of the manifold's individual states. The same argument allows the separation of the spin and orbital angular momentum belonging to the $3p^4(^1D)$ core and 4p outer electron. This is the cornerstone of the separation the subshell multipoles presented in this paper, where we observe the multipoles of several states in a given manifold through the integrated Stokes parameters.

In this case there are only three independent parameters to describe the orbital angular momentum distribution :

$$t_{20}(l_c),\ t_{20}(l_o),\ \text{and}\ t_{40}(l_o).$$

Expanding eq. (2) in terms of L, l_c, l_o, and these parameters we find that

$$\begin{aligned} t_{KQ}(L) &= F_L\left(t_{20}(l_c), t_{20}(l_o), t_{40}(l_c)\right) \\ &= \frac{c_{2L}t_{20}(l_c)+c_{3L}t_{20}(l_o)+c_{4L}t_{20}(l_c)t_{20}(l_o)+c_{5L}t_{40}(l_c)t_{20}(l_o)}{1+c_{1L}t_{20}(l_c)t_{20}(l_o)}, \end{aligned} \tag{3}$$

where the coefficients $c_{1L}\ldots c_{5L}$ depend only on L, and the function $F_L()$ is defined for use in the following section.

DATA ANALYSIS

Figure 1a presents the "raw" values of $t_{KQ}(L)$ for the states of interest. Due to the non-linear nature of eq. (3), some care must be taken in determining the values of the $t_{KQ}(L)'s$. To address this issue we inverted the equation using a terrain search

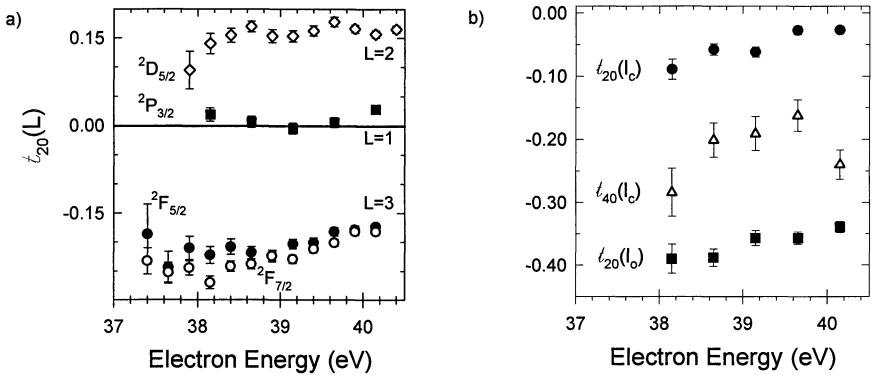

Figure 1a) The multipoles of the total angular momentum, L, generated from the relative Stokes parameters of the resonance fluorescence due to the reaction (1). **Figure 1b)** The calculated values of the multipoles of the $3p^4(^1D)$ and 4p subshells of the Ar^+ ion excited by electron impact.

algorithm and computed the uncertainties associated with the derived quantities (the multipoles of l_i and l_o) using a Monte-Carlo method.

The terrain search algorithm minimizes the Euclidian distance, d, between the measured vector $\{t_{20}(L=3),\ t_{20}(L=2),\ t_{20}(L=1)\}$ and the estimated vector $\{F_3(t_{20}(l_c),t_{20}(l_o),t_{40}(l_o)),\ F_2(t_{20}(l_c),t_{20}(l_o),t_{40}(l_o)),\ F_1(t_{20}(l_c),t_{20}(l_o),t_{40}(l_o))\}$, weighted by the uncertainty in the measured values. To find the global minimum of d it is generally necessary to find the smallest local minimum in d by starting at various points in $\{t_{20}(l_c),\ t_{20}(l_o),\ t_{40}(l_o)\}$ space. Fortunately, because they are derived from angular momenta, the space of $\{t_{20}(l_c),\ t_{20}(l_o),\ t_{40}(l_o)\}$ is bounded by a finite region so it is possible to search the space exhaustively or by the more efficient method of random sampling.

Though the propagation of errors technique is the *de facto* standard for computing the uncertainty in derived quantities, given a known uncertainty in the data, this method has some significant drawbacks. It can give misleading results if the function that must be evaluated has high curvature in the region of interest. However it is possible to use Monte-Carlo methods to determine these uncertainties without any prior knowledge or prior assumptions about the function that must be determined [13]. The method relies on generating an artificial set of points that stands in as a proxy for a given datapoint and its uncertainty. Therefore the artificial set must encompass all *a priori* knowledge of data : i.e. it must be statistically indistinguishable from the parent distribution from which the data was drawn. Given the fact that we can derive values and uncertainties for the $t_{20}(L)$ from experimental data with known statistical properties, we know that the Gaussian distribution corresponds very closely to the actual parent distribution of each of the $t_{20}(L)$.

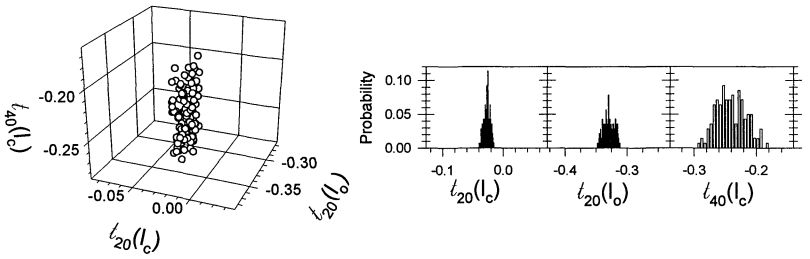

Figure 2 A representation of the distribution of the multipoles of the $3p^4(^1D)$ core and 4p outer electron generated from the set of synthetic data corresponding to the 40.2eV datapoint of Fig. 1a.

Figure 2 is an example of the typical "solution cloud" that we obtained for each energy we investigated. The top graph is a scatter plot of the local minima for the simulated set of data at an incident electron energy of 40.2 eV. The bar graphs below show the projection of this probability distribution onto the three individual axes. The mean value and width of these distributions corresponds to the solution and uncertainty in the quantities $\langle t_{20}(l_c)$, $\langle t_{20}(l_o)$, and $\langle t_{40}(l_c)$. Each graph is unimodal, compact, and could be well described by a Gaussian function. All of these considerations point to the fact that this is a good, well defined solution.

With only moderate assurance that the RB hypothesis holds for the core and orbital angular momenta, we were somewhat concerned that the multipoles might not factor as in eq. (3), i.e. that are $\langle t_{20}(l_c)$ and $\langle t_{20}(l_o)$ are correlated. In this case there would be only three measurements and four independent parameters :

$$t(l_c,l_o)_{20,00},\ t(l_c,l_o)_{00,20},\ t(l_c,l_o)_{20,20},\ \text{and}\ t(l_c,l_o)_{40,20}.$$

If there was correlation between the two parameters $\langle t_{20}(l_c)$ and $\langle t_{20}(l_o)$, then the quantity

$$x = \frac{t(l_c,l_o)_{20,20} - t(l_c,l_o)_{20,00}\, t(l_c,l_o)_{00,20}}{t(l_c,l_o)_{20,20} + t(l_c,l_o)_{20,00}\, t(l_c,l_o)_{00,20}} \qquad (4)$$

which measures this correlation, would be non-zero. Figure 3 presents the solution of the listed multipoles for many choices of x. It is obvious that the quoted value of $\langle t_{40}(l_c) \sim \langle \mathcal{T}^{\dagger}_{40}(l_c) \otimes \mathcal{T}^{\dagger}_{20}(l_o) \rangle / \langle \mathcal{T}^{\dagger}_{00}(l_c) \otimes \mathcal{T}^{\dagger}_{20}(l_o) \rangle$ is consistent with the graph up to x = 0.5. In combination with the fact that the RB hypothesis appears to hold for the orbital angular momentum, the results in Figure 3 indicate that our analysis can accurately determine the hexadecapole of the $Ar^+ 3p^4(^1D)$ core.

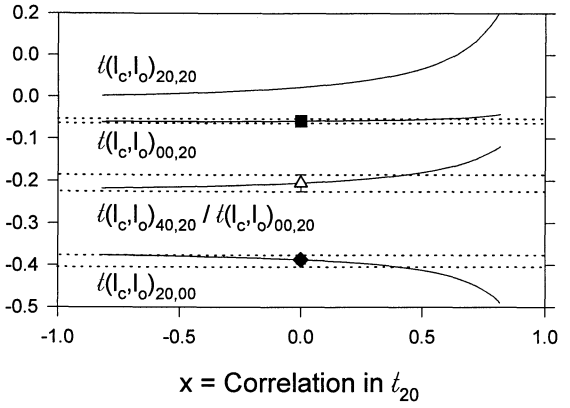

Figure 3 The calculated values of the multipoles of the $3p^4(^1D)$ core and outer 4p electron, as a function of correlation (eq. (4)). The points correspond to the reported values assuming no correlation, and the dotted lines correspond to the 1σ error bars.

RESULTS AND CONCLUSIONS

Applying the method described in the previous section we were able to deduce values of the multipoles for the $3p^4(^1D)$ core and 4p outer electron subshells. These subshell multipoles are presented in Figure 1b. This method hinges on the applicability of the RB hypothesis. The measurements reported here would not be invalidated, even if the RB hypothesis did not hold rigidly true for the core/outer electron subsystems. The similarity of the $t_{20}(L=2)$ multipoles for J = 5/2 and 7/2 in (Figure 1a) confirms this for fine-structure relaxation, as do our spin polarized measurements [11].

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