Shakeoff measurement of the $l=3$ states of barium

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Isolated core excitation was used to produce low-energy continuum electrons in the $l=3$ angular momentum state of barium. Data were taken over a region of energy that coincided with the energy of the $6p_{3/2}nf$ doubly excited states. Analysis of the data using multichannel quantum defect theory allowed the measurement of the widths of the $6p_{3/2}nf$ states and the energy-dependent phase of the continuum electronic wave functions due to interaction with the doubly excited states. The phase of the continuum electrons is shown to vary continuously with energy, due to the anomalously broad widths of the double excited states. [S1050-2947(98)02809-1]

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INTRODUCTION

In a previous experiment, a method was developed that can measure the phase of low-energy continuum electrons in well-defined angular momentum states, due to interaction with doubly excited states [1]. This method, which uses an isolated core excitation [2] (ICE) to produce a low-energy continuum electron in the presence of an excited ionic core, is equivalent to a measurement of the phase shift of an electron scattering off the ion. Because the electron is in a well-defined angular momentum state, the measurement gives the phase of individual partial waves. In the present work, this technique is applied to the $l=3$ angular momentum states. These states are of particular interest because of the anomalously broad widths of the $6p_{3/2}nf$ states [3,4]. The widths of these states is explained by the fact that the inner classical turning point of the $l=3$ Rydberg electron coincides with the outer classical turning point of the electron in the $6p$ ionic state, producing a large interaction between the two electrons.

In a normal spectroscopic measurement of the widths of autoionizing states using the ICE method, the widths of the states can be directly obtained from the excitation cross sections. In the case of the $6p_{3/2}nf$ states whose widths are a significant fraction of the spacing between states, the excitation cross sections are dominated by the energy-dependent overlap between the initial and final Rydberg states [5], which reduces the accuracy of the width measurements. In the present work, a shakeoff process is used to directly measure the interchannel couplings between the bound and continuum channels. This measurement, in addition to providing the phase of the continuum electron wave function, gives an accurate measurements of the widths of the autoionizing states.

EXPERIMENT

Four tunable, pulsed dye lasers were used to excited barium as shown in Fig. 1. The dye lasers were focused and intersected in the interaction region inside a vacuum chamber. The dye lasers, which were all linearly polarized in the vertical axis, were pumped with either the second or third harmonic of a Nd:YAG laser. The Nd:YAG laser operated at 20 pulses/sec, with a pulse duration of 10 ns. Barium atoms were produced using a resistively heated, effusive oven. The resultant atomic beam was collimated using a 1-mm aperture and crossed the dye laser beams at right angle in the interaction region. The first three lasers were used to excite the atoms from the $6s^2\ ^1S_0$ ground state through the $6s6p\ ^1P_1$ state and the $6s6d\ ^1D_2$ state, to the $6s21f\ ^1F_3$ state. After a delay of approximately 70 ns, which allowed the decay of the $6s6d\ ^1D_2$ intermediate state, the fourth laser, which was amplified using a two-stage dye amplifier, was used to excite the ionic core of the barium atoms. The wavelength of the fourth laser was scanned from 453 to 495 nm. This range included both the $6s$ to $6p_{1/2}$ ionic resonance (493 nm) and the $6s$ to $6p_{3/2}$ ionic resonance (456 nm).

Figure 2 shows the signal, which consists of all electrons produced by the fourth laser, either by autoionization of the doubly excited states or by shakeoff of the Rydberg electron during the core excitation. The electrons were detected by applying a small voltage, using capacitor plates located above and below the interaction region, which swept the electrons through a screen mess in the top plate into a charge particle detector. The signal produced by the detector was

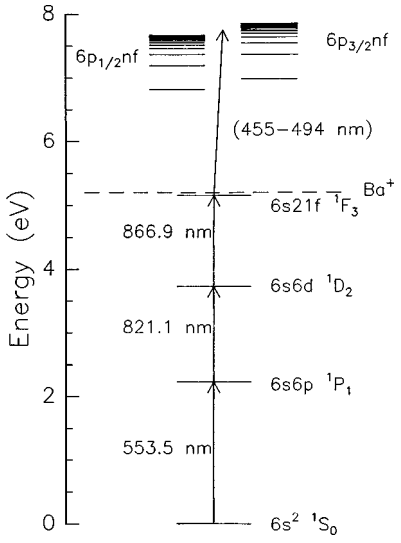

FIG. 1. Four photon excitation of barium is shown with the relative atomic states. Three lasers were used to excite the $6s21f\ ^1F_3$ state. The fourth laser was scanned over the energy region, which includes the $6p_{1/2}$ and $6p_{3/2}$ ionic states.

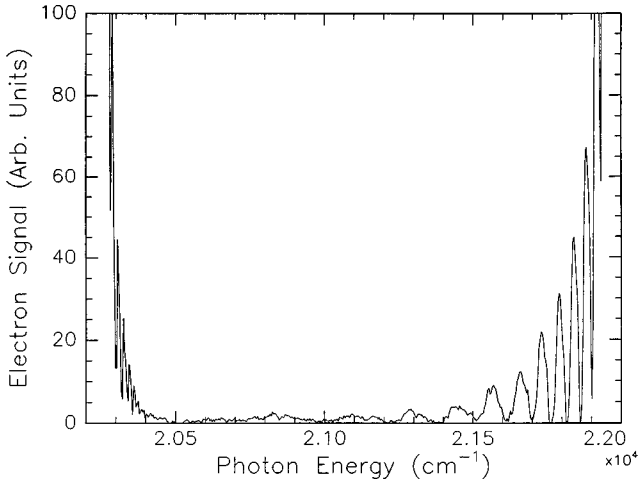

FIG. 2. The electron signal is shown vs the energy of the fourth photon. The large structures at either end of the scan is due to shakeup excitation of the $6p_{1/2}nf$ and $6p_{3/2}nf$ states. Shakeoff into the $6p_{1/2}\epsilon f$ continuum occurs at photon energies above 20 522 cm^{-1}.

averaged by a digital oscilloscope and stored on a computer. The large features in the data correspond to shakeup transitions near the $6s$ to $6p_{1/2}$ and $6s$ to $6p_{3/2}$ ion resonances. For photon energies above 20 522 cm^{-1} a shakeoff excitation could occur, corresponding to the $6s21f$ to $6p_{1/2}\epsilon f$ transition. In the region directly between the two ionic resonances the data represents a combination of shakeup to the $6p_{3/2}nf$ states and shakeoff to the $6p_{1/2}\epsilon f$ continuum.

THEORY

The binding energy of an electron in a bound state of an atom, in atomic units, is given by

$$W=-\frac{1}{2(n-\delta)^2}=-\frac{1}{2(n^*)^2}, \tag{1}$$

where n is the principal quantum number, δ is the quantum defect, which depends on the angular momentum of the electron and the ionic core configuration, and n^* is the effective quantum number of the electron. In the case of an isolated Rydberg series, the quantum defect is independent of n, at least for the relatively high-n states, so that a single quantum defect can be used to describe the entire Rydberg series. The phase of the bound electronic wave function, relative to hydrogen, is given by $\tau=\pi n^*$. This phase is a measure of the relative contributions of regular and irregular Coulomb wave functions, which make up the energy eigenstates [6,7]. Above the ionization limit where electron energies form a continuum, the relative phase of the electron can still be described by τ using the definition $\tau=-\pi\delta$. For an isolated continuum, at least at relatively low electron energies, δ is independent of energy and equal to the quantum defect of the bound Rydberg series below the ionization limit. The lack of energy dependence of the quantum defect is due to the fact that the small r electronic wave function is relatively insensitive to small changes in energy. If the continuum is coupled to a series of doubly excited states, then the phase of the continuum varies with energy. Specifically, the phase of the continuum shifts by π as the energy transverses the position of a doubly excited state [1]. An example of this phase variation occurs in barium in the energy range between the $6p_{1/2}$ and $6p_{3/2}$ ionic states where $6p_{1/2}\epsilon l$ continuum states interact with $6p_{3/2}nl$ doubly excited states.

The variation of the electron phase can be measured directly by using ICE technique to perform a shakeoff transition. In this process an atom is initially prepared in a singly excited Rydberg state. A laser is then tuned slightly above the $6s$ to $6p_{1/2}$ ionic transition to excite the ionic core electron. The transition moment for this excitation is given by [5]

$$T=\mu_{1/2}A_{\epsilon l}\frac{\sin(\tau_{\epsilon l}-\tau_i)}{W_{\epsilon l}-W_i}, \tag{2}$$

where $\mu_{1/2}$ is the $6s$ to $6p_{1/2}$ dipole moment, $A_{\epsilon l}$ is the density of final states that is a constant for the continuum states. The third term in the expression represents the overlap between the initial Rydberg state and the final continuum state, where the initial electron phase and binding energy are given by τ_i and W_i, respectively. For a shakeoff transition in which the final state of the electron is a continuum state $W_{\epsilon l}$ is positive and the variation of $\tau_{\epsilon l}$ with energy is due only to interaction with doubly excited states. The energy denominator is a measure of the detuning from the $6s$ to $6p_{1/2}$ ionic transition.

In the region of interest, between the two ionic transitions, there is also the possibility of a shakeup excitation to the $6p_{3/2}nf$ states. The full transition moment is given by the sum of both the shakeoff and shakeup transitions.

$$T=\mu_{1/2}A_{\epsilon l}\frac{\sin(\tau_{\epsilon l}-\tau_i)}{W_{\epsilon l}-W_i}+\mu_{3/2}A_{nl}\frac{\sin(\tau_{nl}-\tau_i)}{W_{nl}-W_i}, \tag{3}$$

where $\mu_{1/2}$ and $\mu_{3/2}$ represent the $6s$ to $6p_{1/2}$ and $6s$ to $6p_{3/2}$ dipole moments, respectively, and the energy denominators represent the detunings from the respective ionic transitions. Note that τ_{nl} is the phase of the $6p_{3/2}nf$ bound states and depends on the binding energy of the Rydberg electron.

The density of states and wave-function phases can be calculated using multichannel quantum defect theory (MQDT). The formalism used to model this experiment is derived elsewhere [8,9] and will be only briefly described here. A three-channel model will be used that includes the $6p_{3/2}nf$ bound channel, the $6p_{1/2}\epsilon f$ continuum channel, and a third channel, which represents all other continua. The eigenstates of the system can be found by diagonalizing the eigenequation

$$[R+\tan(\tau_m(E)+\pi\delta_m)]a=0, \tag{4}$$

where the tangent terms represent a three by three diagonal matrix with m denoting the channel and a representing the eigenstate. R is a three by three matrix with zero diagonal elements and off diagonal elements, which represent the interchannel couplings. In this experiment no information is obtained regarding the continuum-continuum coupling so that this coupling is set to zero. c_1 will be used to represent the coupling between the $6p_{3/2}nf$ and $6p_{1/2}\epsilon f$ channels and c_2 will represent the coupling between the $6p_{3/2}nf$ and all

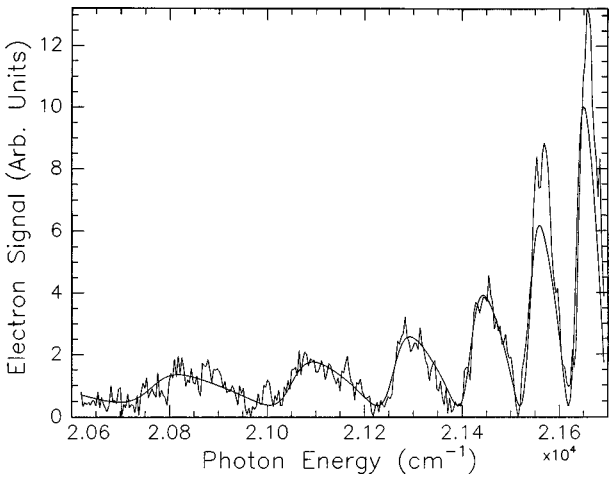

FIG. 3. Blown up view of the energy region in which shakeoff contributes to the excitation cross section is shown. In the lower-energy region both shakeup and shakeoff processes contribute to the signal. At higher-energy shakeup excitation dominates the spectra. The MQDT fit of the spectra is shown as a smooth line along with the data.

other continua channels. Diagonalization of Eq. (4) gives two eigenstates, one of which is decoupled from the bound channel.

$$|+\rangle = A^{+}_{6p_{1/2}\epsilon f}|6p_{1/2}\epsilon f\rangle + A^{+}_{\epsilon}|\epsilon\rangle + A_{6p_{3/2}nf}|6p_{3/2}nf\rangle \quad (5)$$

and

$$|-\rangle = A^{-}_{6p_{1/2}\epsilon f}|6p_{1/2}\epsilon f\rangle + A^{-}_{\epsilon}|\epsilon\rangle, \quad (6)$$

where ϵ represents the continua other than the $6p_{1/2}\epsilon f$ continua. The relevant densities of states are given by

$$A^{+}_{6p_{1/2}\epsilon f} = \frac{c_1}{\sqrt{c_1^2+c_2^2}} \quad \text{and} \quad A^{-}_{6p_{1/2}\epsilon f} = \frac{-c_2}{\sqrt{c_1^2+c_2^2}} \quad (7)$$

for the continuum and

$$A_{6p_{3/2}nf} = -\left(\frac{(c_1^2+c_2^2)[1+\tan^2(\tau_{nf}+\pi\delta_f)]}{(c_1^2+c_2^2)^2+\tan^2(\tau_{nf}+\pi\delta_f)}\right)^{1/2} \quad (8)$$

for the bound states, where δ_f is the quantum defect of the unperturbed $6p_{3/2}nf$ channel. The ϵf electron phase for the purely continuum eigenstate is constant and simply given by $\tau^{-}_{\epsilon f} = -\pi\delta_{\epsilon f}$, where $\delta_{\epsilon f}$ is the quantum defect of the unperturbed $6p_{1/2}\epsilon f$ channel. The electron phase of the eigenstate containing bound character is given by

$$\tau^{+}_{\epsilon f} = \arctan\left(\frac{c_1^2+c_2^2}{\tan(\tau_{nf}+\pi\delta_f)}\right) - \pi\delta_{\epsilon f}. \quad (9)$$

the total cross section is proportional to the sum of the squares of the transition moments to both of the eigenstates:

$$\sigma \propto (T^{+})^2 + (T^{-})^2.$$

The T^{+} term contains the sum of both the transition to the bound and continuum channels as seen in Eq. (3). The T^{-} term contains only shakeoff, since the $|-\rangle$ state contains no bound character. The only energy dependents of the T^{-} term is due to the detuning from the ion resonance.

TABLE I. The unperturbed quantum defects of the $6p_{3/2}nf$ channel and $6p_{1/2}\epsilon f$ channel are shown along with the interchannel couplings, with c_1 being the $6p_{3/2}nf$-$6p_{1/2}\epsilon f$ coupling and c_2 being the coupling between the $6p_{3/2}nf$ channel and all other continuum channels.

$\delta_{6p_{3/2}nf} = 0.318 \pm 0.05$
$\delta_{6p_{1/2}\epsilon f} = 0.444 \pm 0.04$
$c_1 = 0.037 \pm 0.015$
$c_2 = 0.774 \pm 0.05$

RESULTS

Figure 3 shows data in the region just above the $6p_{1/2}$ ionization limit. The higher-energy region of the data is dominated by shakeup to the $6p_{3/2}nf$ states. The lower-energy region of the data has contributions from both shakeup and shakeoff to the $6p_{1/2}\epsilon f$ continuum. The smooth curve shown along with the data is a fit using the couplings between the $6p_{3/2}nf$ channel and the two continua, along with the quantum defects of the $6p_{3/2}nf$ channel and the $6p_{1/2}\epsilon f$ channel as fit parameters. The values obtained for these parameters are listed in Table I. The shakeoff excitation contributes to the excitation spectra in two ways. The uncoupled $6p_{1/2}\epsilon f$ continuum produces a signal that decreases as the square of the detuning from the $6s$ to $6p_{1/2}$ ionic transition. This aspect of the signal is small for the f states since the quantum defect of the unperturbed $6p_{1/2}\epsilon f$ continuum is nearly the same as that of the bound $6snf$ states; 0.444 and 0.471, respectively. Because of this small change in quantum

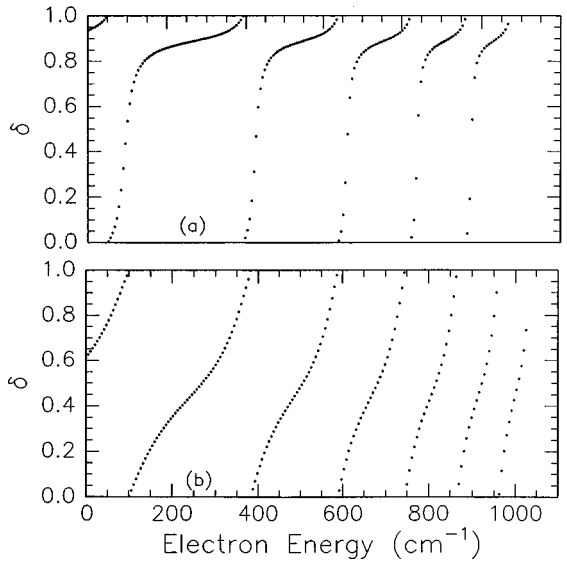

FIG. 4. Energy-dependent quantum defects are shown for (a) the $6p_{1/2}\epsilon d$ states and (b) the $6p_{3/2}\epsilon f$ states. Only the noninteger value of the quantum defects is shown. In the case of the d states the variation occurs over a small energy region. Between the regions of rapid variation the quantum defects are approximately equal to the unperturbed value of 2.89. In the case of the f states, the variation occurs continuously, due to the large widths of the doubly excited states.

defect, the overlap between the bound Rydberg state and the unperturbed continuum is extremely small, leading to a small transition moment to the $|-\rangle$ continuum state. The contribution to the excitation spectra from shakeoff to the $|+\rangle$ state is also rather small due to the small coupling between the $6p_{1/2}\epsilon f$ and $6p_{3/2}nf$ channels. This is not the ideal situation since one would like the spectra to be dominated by the shakeoff spectra to achieve the most ideal measurement. However, the contribution of shakeoff to the total excitation spectra was sufficient to determine the parameters shown in Table I. The relatively small coupling between the $6p_{1/2}\epsilon f$ and $6p_{3/2}nf$ channels is interesting in that it suggests that the $6p_{3/2}nf$ states autoionize primarily to high-energy continua.

With the values listed in Table I, the energy-dependent phase of the $6p_{1/2}\epsilon f$ electronic wave function can be calculated using Eq. (9). Figure 4 shows a plot of the quantum defect of the $6p_{1/2}\epsilon f$ channel versus electron energy, which is related to the phase by $\tau=-\pi\delta$. Also shown, for comparison, is the quantum defect of the $6p_{1/2}\epsilon d$ channel calculated from a previous measurement [1]. The figure shows that, in the case of the d states, δ_d varies rapidly over a small energy region at the locations of the doubly excited states. Between these regions of rapid change, δ_d is relatively constant and equal to the unperturbed quantum defect of the $6p_{1/2}\epsilon d$ channel. The f state quantum defect, on the other hand, varies continuously and nearly uniformly over the entire energy range. The continuous variation of δ_f is due to the anomalously broad widths of the $6p_{3/2}nf$ states.

The widths (Γ_n is the full width at half maximum in atomic units) of these states can be calculated from the density of states, Eq. (8), using the measured values of the interchannel couplings.

$$\Gamma_n=\frac{2(c_1^2+c_2^2)}{\pi n^3}=\frac{0.382}{n^3}.$$

Since the spacing between adjacent Rydberg states is $1/n^3$, in atomic units, this width represents 0.382 times the spacing between state. Because of the broad widths of these states, the bound state character of the f states is a series of overlapping resonances. It is this overlap that makes a normal width measurement difficult, which is necessary to determine the interchannel couplings. In the present measurement, which directly measures the phase of the final-state continuum wave function, the problems associated with the overlapping resonances are overcome.

ACKNOWLEDGMENTS

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