Experimental determination of the total angular momenta of autoionization states in multistep photoionization of atoms

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The total angular momenta of even autoionization resonances of the $6p7p$ configuration have been determined experimentally through the use of various combinations of the polarizations of the radiation in a two-step photoionization of $Ba(^1S_0)$ atoms.

Autoionization states have been studied in the optical absorption spectra of atoms for more than fifty years. In nearly all cases, they have been identified on the basis of various theoretical considerations such as the Hund role, the analogy with lines in the discrete spectrum, and, just recently, a direct comparison with the results of various calculations.[1–3] At the moment, however, the calculation accuracy is not good enough for identifying autoionization states. The experimental identification methods based on the Zeeman and Stark effects which are used in the discrete spectrum cannot be used for autoionization states, since the widths of the resonances are far greater than their splitting in the fields attainable in the laboratory.

There are, on the other hand, some other possibilities for experimentally identifying autoionization states. During single-photon ionization, one could measure the degree of polarization of the photoelectrons.[4] During multistep ionization, it would be simpler to use various combinations of polarized light in the different steps. Another possibility, suggested by Bekov *et al.*,[1] would be to excite autoionization states through various intermediate states.

Let us consider as an example the particular case of the two-step ionization of an atom with a zero total angular momentum in its initial state through an intermediate resonance. Selection rules for the absorption of each of the photons (in the dipole approximation) follow from the condition under which the $3j$-symbol

$$\begin{pmatrix} J' & 1 & J_0 \\ -M' & \lambda & M_0 \end{pmatrix},$$

which appears in the amplitude for the process,[5] is not equal to zero. Here J_0,M_0 and J',M' are the total angular momentum and its projection for the initial and final states, respectively; and λ is the spin projection of the photon, having the value $\lambda = 0$ for a linearly polarized photon and $\lambda = \pm 1$ for a circularly polarized photon. Another important point is that the quantization axes for linearly and circularly polarized photons are different: For linear polarization, the quantization axis coincides with the polarization vector, while for circular polarization it coincides with the direction of the (vector) momentum of the photon.

Using the well-known conditions under which the $3j$-symbols are nonzero,[5] we find that for certain combinations of the polarizations in the different steps it would be possible to excite autoionization states with only certain values of the total angular momentum J. Specifically, (1) if the laser beams used in both steps are linearly polarized in the same direction, autoionization states with $J = 0$ and $J = 2$ will be excited; (2) if the beams from the lasers in both steps are linearly polarized, but in mutually perpendicular directions (since the quantization axes for such photons do not coincide, the wave function of one must be expanded in basis function in a rotated coordinate system), autoionization states with $J = 1$ and $J = 2$ will be excited; and (3) if the beams from the lasers in the two steps are circularly polarized, in the same direction, only autoionization states with $J = 2$ will be excited.

In the second case one could use a combination of linearly and circularly polarized beams under the condition that the photon beams are directed at right angles with respect to each other, so that their quantization axes coincide.

By establishing that an autoionization state is not present experimentally for certain combinations of the polarizations, one could unambiguously determine its total angular momentum J. The same method can be used to determine the total angular momenta of discrete excited states.

This method will obviously also be applicable in the case in which there are more than two steps. If a ground-state atom has a nonzero total angular momentum, it would be necessary to set up the initial state for this method in such a way that only states with a certain projection of the total angular momentum are populated. This situation can be achieved by means of optical pumping or by the Stern-Gerlach method. Another possibility is to choose a sequence of steps such that the atoms in one of the intermediate states have definite values of the total angular momentum and of its projection.

The method described above has been used to determine the total angular momenta of even autoionization states in Ba atoms corresponding to the $6p7p$ configuration and lying in the energy interval 49 000–54 500 cm^{-1}. The atoms are excited by two dye lasers, pumped simultaneously by a common Nd:YAG laser. The interval over which the wavelength is tuned is 540–640 nm for one of the lasers, with a power

TABLE I. Values found for the total angular momenta of autoionization states in the present study.

E, cm^{-1}, exptl.[2]	Identification proposed in Ref. 2		Present study
50 383	$^1P_1$	$(1/2, 1/2)_1$	2
51 113	$^3D_1$	$(1/2, 3/2)_1$	2
51 491.5	$^3P_0$	$(1/2, 1/2)_0$	2
52 158	$^3P_1$	$(3/2, 1/2)_1$	1
52 583	$^3P_2$	$(3/2, 1/2)_2$	1

of 10 kW in the pulse, while that for the other is 270–320 nm, with a power of 2 kW. The spectral width of the output line is 3 cm^{-1}. The autoionization states are excited through $6snp\,^1P_1$, $n = 6, 7, 8$, intermediate levels. The laser beams intersect the beam of atoms between the grids of the ion source of a time-of-flight mass spectrometer. The ion current is measured with the help of a stroboscopic voltage transformer. Of the eight autoionization states which have been observed previously[2] we studied five resonances. The results of the measurements of the total angular momenta are shown in Table I, along with data from Ref. 2. For four of the autoionization states, the results of our measurements contradict the identification proposed in Ref. 2 on the basis of a comparison with calculations. The recently published[6] calculations in the Hartree-Fock-Dirac approximation, which allow for the superposition of configurations, yield results quite different from the results calculated in Ref. 2. The very order of the autoionization states is different.

Consequently, the only reliable way to identify autoionization states at present is experimentally, and the method proposed in the present letter is a simple method, applicable to essentially any atom.

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