Relativistic and Interchannel Coupling Effects in Photoionization Angular Distributions By Synchrotron Spectrocopy of Laser Cooled Atoms

L. H. Coutinho,[1,2] R. L. Cavasso-Filho,[1,*] T. C. R. Rocha,[1,2] M. G. P. Homem,[2] D. S. L. Figueira,[1] P. T. Fonseca,[2] F. C. Cruz,[1] and A. Naves de Brito[2,3,†]

[1]*Instituto de Física "Gleb Wataghin," Universidade Estadual de Campinas (UNICAMP), Box 6165 CEP 13083-970 Campinas-SP, Brazil*

[2]*Laboratório Nacional de Luz Síncrotron (LNLS), Box 6192 CEP 13084-971 Campinas-SP, Brazil*

[3]*Instituto de Física, Universidade de Brasília (UnB), Box 4455 CEP 70910-900 Brasília-DF, Brazil*

(Received 10 December 2003; published 25 October 2004)

We investigate the angular distribution of photoionization fragments at low photon energies (12–40 eV) in an open shell atom, by synchrotron radiation recoil ion momentum spectroscopy in a laser cooled and trapped sample. For cesium atoms, for which relativistic effects play an important role and the ion recoil is relatively small, we could determine large and rapid changes of the asymmetry parameter β from two, observed for s electrons outside resonances and far from the Cooper minimum. They can be explained by relativistic effects and interchannel coupling arising from final state configuration mixing.

DOI: 10.1103/PhysRevLett.93.183001 PACS numbers: 32.80.Fb, 32.80.Pj

Complex problems, such as many-body interactions or relativistic effects at atomic scales, are better studied in simple and isolated systems interacting or "colliding" with photons. In particular, the interaction of ionizing radiation with an atom provides an excellent "laboratory" for the study of the complex electronic dynamics. In photoionization, the electromagnetic radiation causes a small perturbation on the atom [1], allowing the unambiguous study of the atomic target [2]. The ideal of studying unperturbed atoms at rest, on the other hand, has been very much approached by laser cooling and trapping techniques [3]. They greatly impacted different areas such as quantum optics, atomic spectroscopy, and metrology, and at the same time have been essential for producing quantum degenerate gases [4,5], which in turn promise to shine light into complex many-body problems involved in superconductivity and superfluidity [6]. The fundamental interaction between photons and atoms also provides the most stringent tests to *ab initio* calculations. The acquired knowledge is often used to model more complex systems. On the experimental aspect of atomic photoionization, measurements of total cross sections have been the most valuable source of information [2]. However, there is a need for alternative and more sensitive techniques. For example, total cross sections depend on the square of transition amplitudes to the various possible final states [7–9], while the photoelectron angular distributions (differential cross sections) depend not only on ratios of amplitudes but also their relative phases, thus allowing much more information to be obtained in principle. For example, it is well known that near ionization thresholds relativistic effects are responsible for dramatic changes in the angular distribution (Cooper minimum [10]), where there is no significant variation in the total cross section. Recent theoretical predictions indicate the presence of another class of minima due to many-body effects [interchannel coupling (IC) [8]]. This is relevant because IC is found to be important for the majority of atoms, subshells, and energies [8,10]. The most advanced techniques have not been able to confirm this prediction so far. Here we present strong indications of their existence. For this we developed a new experimental technique to measure the photoelectron angular distribution, which combines laser cooling and trapping with synchrotron radiation ion momentum spectroscopy [11]. We have used cesium atoms for which relativistic and many-body effects (electron-ion interactions) are important and the ion recoil is small. This new technique applied to an open shell one-electron atom with large mass, such as cesium, allowed precise off-resonance measurements of the photoionization angular distribution parameter β. They revealed strong deviations of β from the nonrelativistic value of 2, for s electrons, at energy regions where the total cross section is completely smooth, and far from the Cooper minimum [10]. They can be explained by quantum interference among final state wave functions (interchannel coupling) [8], observed here for the first time.

For linearly polarized ionizing radiation in the dipole approximation [12], the photoelectrons have an angular dependence determined by the β parameter. The differential cross section is given by

$$\frac{d\sigma(h\nu)}{d\Omega} = \frac{\sigma(h\nu)}{4\pi}\left[1 + \beta(h\nu)\frac{3\cos^2\theta - 1}{2}\right]$$

where θ is the angle defined by the directions of the polarization vector ($\hat{e}$) and the photoelectron and σ is the total cross section. The asymmetry parameter β can vary in the range from 1 to 2, determined by the requirement that the cross section be non-negative. For ioniza-

tion of ns electrons from closed shell atoms β is always equal to two nonrelativistically. In the vicinity of a Cooper minimum [10], owing to relativistic spin-orbit effects which are more important for high Z elements, β can suffer strong variations because instead of only one $s \rightarrow p$ partial wave, two waves with differing amplitudes are now present: $s \rightarrow p_{1/2}$ and $s \rightarrow p_{3/2}$. The Cooper minimum for cesium [10], for example, is located in a region near 1.6 eV above the $6s$ ionization threshold. Near autoionizing resonances, dipole amplitudes can have slightly different energy dependences, causing β to rapidly change with energy [8]. For open shell atoms and ns shell photoionization, a more complex situation is present, since the ion plus photoelectron state is often characterized by more than one term and differences in the amplitudes of these terms can make β energy dependent [8]. Here we confirm β variations due to coupling between spin-orbit split channels.

Although photoionization of open shell atoms may reveal a rich set of interesting phenomena, there are relatively few experimental studies for them, with much fewer involving differential cross sections. The main reason is connected to the experimental challenges. They often need to be heated before it is possible to reach a useful partial pressure near 10^{-6} mbar. One way to measure differential cross sections for open shell atoms is to use two-electron analyzers [13]. This method allows high resolution for electron kinetic energy and is an excellent tool to obtain β at autoionizing resonances. However, only the fraction of electrons emitted in the direction of the detector within a small solid angle are collected. Often, for open shell atoms the characteristic low-density atomic beams do not allow proper determination of β outside resonances where interesting effects, such as interchannel coupling could be detected. One way to obtain complete angular detection efficiency over 4π is to use cold targets and measure the recoil of the created ion, thus extracting the angular distribution of the ejected photoelectrons. So far this has been done by cold target recoil ion momentum spectroscopy (COLTRIMS) [14] by using supersonic beams for 1D velocity compression. Nevertheless, the supersonic beam technique is restricted to closed shell gases because several bars of pressure are needed in the back of a needled inlet system to achieve supersonic beam speed. This represents several orders of magnitude higher pressure than currently is possible to obtain with most open shell atoms. In addition, thermal broadening reduction in COLTRIMS is currently limited to near 10 μeV [15] and to only one dimension. This rule out the possibility to study larger open shell atoms such as cesium ($Z = 55$), since near a threshold region the ion recoil energy may be as low as 0.5 μeV. More recently, laser cooled samples with 3D velocity reduction have been employed for ion momentum spectroscopy using lasers as the ionizing radiation [16], and for scattering experiments using ion beams [9]. The novel technique presented here overcomes the shortcomings described above. Laser cooling and trapping [3] allows orders of magnitude higher atomic density in the collision center and very low pressure elsewhere. In addition, 3D kinetic energy reduction, down to a few tens of nanoelectronvolt, is obtained in a standard way.

The experiment was performed at the Brazilian synchrotron light source (LNLS). We have used laser cooled and trapped cesium atoms, with thermal energies near 10 neV, and a synchrotron light source operated in single-bunch mode. We employed a one-meter long time-of-flight (TOF) spectrometer, described elsewhere [11]. The ions arrive in the detector, 25 mm in diameter, within a spot smaller than 1 mm. Figure 1 shows a schematic diagram of the experimental setup, with the magnetic optical trap (MOT) coils and the TOF spectrometer. Differential cross sections can be affected by smaller multipole contributions even at low photon energies [17], but our experimental arrangement with the TOF axis placed in the so-called dipole plane is insensitive to them. Complete information on the differential cross section can be obtained once the geometry and density of the sample are known [17]. However, we will concentrate here on the asymmetry parameter β. The flight time for Cs^+ ions is about 85 μs, but as our experimental window was determined by the synchrotron pulse period of 311 ns, the arriving ions were summed up in this time interval. The cesium outer valence configuration is given by Core; inner-valence; $4d^{10}5p^6 6s^1$. The binding energies of interest are $6s^{-1} = 3.89$ eV; $5p^{-1}(P_{3/2}) = 17.2$ eV; $5p^{-1}(P_{1/2}) = 17.6$ eV [18].

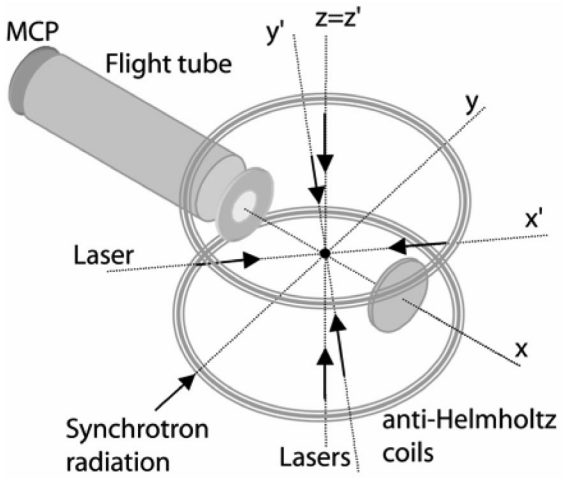

FIG. 1. Schematic view of the experimental setup. The axis of the TOF spectrometer is perpendicular to the propagation direction of the synchrotron UV photons and parallel to their electric field. The six cooling laser beams are located along three orthogonal axis, x', y', and z'. MCP: microchannel plate.

Depending on the β parameter, the time-of-flight spectra in Doppler-free samples show different shapes as shown in Figs. 2 and 3. In Fig. 3 we show three TOF spectra, where circles represent the experimental data, while the solid line is a Monte Carlo simulation of the ions trajectory, including all relevant parameters [19]. From these simulations we obtained the asymmetry parameters and ion recoil energies (IRE). In Fig. 3(a) the spectrum corresponds to photoelectrons ejected only from the $6s$ orbital. The photon excitation energy is indicated in the total ion yield (TIY) spectrum of Fig. 4, which is proportional to the total cross section. The double peak structure is characteristic of an asymmetry parameter larger than zero. Furthermore, the IRE is in good agreement with the excess energy of the ejected electrons. In Fig. 3(b) it is important to notice the very small kinetic energy of $5p$ ejected electrons, which would not be detected by electron spectroscopy due to a strong background from low kinetic energy electrons. Taking into account the photon energy distribution, the spectrum in Fig. 3(b) allows a direct determination of the ratio between ionization cross sections for $5p$ and $6s$ electrons, which was found to be $\sigma_{5p}/\sigma_{6s} = (1.5 \pm 0.5) \times 10^3$. In Fig. 3(c) the obtained asymmetry parameter is in good agreement with a theoretical calculation from Ref. [20].

In Fig. 4, the three TOF spectra in the upper part of this figure were taken at the top of resonant excitations, while the two other are off-resonant. Tentative assignments, based on relativistic Hartree-Fock calculations [21], for these three resonances are $5p^5 6s^{-1} 5d^2 D_{3/2}$ (15.76 eV), $5p^5 6s^{-1} 6d^2 P_{1/2}$ (16.02 eV), and $5p^5 6s^{-1} 6d^2 D_{3/2}$ (16.40 eV). Perhaps the most interesting feature however is connected to the two off-resonant TOF spectra taken at 14.38 and 15.96 eV (Fig. 4). At these energies, where the total cross section (see the TIY in Fig. 4) is smooth, we found $\beta = 0.6 \pm 0.2$ and 0.7 ± 0.2, respectively. These strong departures from $\beta = 2$ at regions outside resonances, where the cross section is small, would involve the presence of a Cooper minimum caused by relativistic spin-orbit effects. This, however, can be discarded since previous calculations show that this minimum lies only 1.6 eV above the $6s$ threshold (3.89 eV) [10] and already at an excitation energy of 12 eV β should be very close to 2. Our observations, nevertheless, are explained by a "second" Cooper minimum induced by IC, predicted for the first time by Altun and Manson [8] and still not observed so far. Because of the possibility of various couplings of the photoelectron with the open shell ionic core, multiple partial waves are possible which interfere with each other, causing differences in the transition amplitudes which make β to differ from two and acquire an energy dependence. This occurs at regions outside resonances where

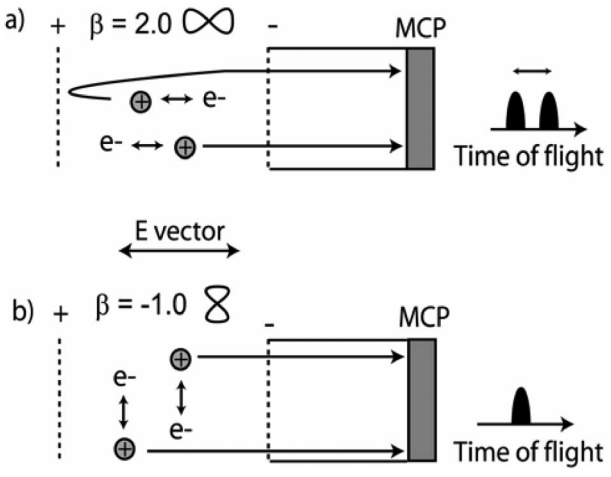

FIG. 2. Effect of the asymmetry parameter β on time-of-flight spectra of cold samples.

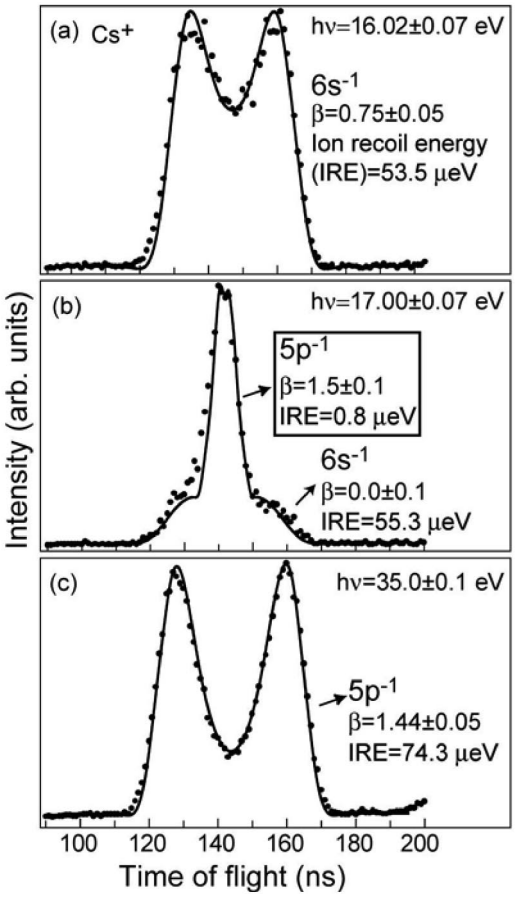

FIG. 3. Typical ion TOF spectra. (a) This photon energy (16.02 eV) corresponds to a strong resonant process involving the excitation of a $5p$ electron to the $6d$ orbital and assigned as $5p^5 6s^{-1} 6d^2 P_{1/2}$; (b) spectrum obtained at 17.00 eV near the Cs $5p$ ionization threshold. Electrons ejected from $5p$ orbital carry much smaller kinetic energy, accounting for the narrow peak. A contribution from $6s$ electrons is clearly identified, giving rise to the pedestal; (c) at 35 eV photoelectrons are ejected mainly from the $5p$ orbital, since in this case the cross section for $6s$ electrons is much smaller [25].

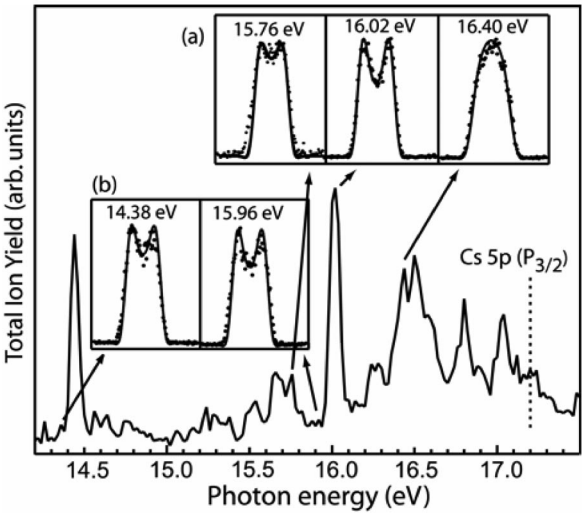

FIG. 4. TIY spectrum with the $5p$ ionization threshold marked. At 15.76 eV a small positive asymmetry parameter ($\beta = 0.3 \pm 0.1$) was found. The other two resonances correspond to the same orbital main configuration but with the ion belonging to different states. The ejected electrons partial wave symmetry are $\kappa s\ ^2P$, $\kappa d\ ^2P, D, F$, for the $^2P_{1/2}$ ionic state and $\kappa s\ ^2D$, $\kappa d\ ^2P, D, F$ for the $^2D_{3/2}$ state, and we have obtained $\beta = 0.75 \pm 0.05$ and $\beta = -0.4 \pm 0.1$, respectively. See text for discussion on the two off-resonant TOF spectra.

the total cross section can be completely smooth. A possible explanation would involve many-body excitation to a partial wave $kd\,^2P, D, F$. It is worth noting that interchannel coupling [8] has already been tried to be confirmed in Ref. [22] with no success. The authors used electron spectrometry to look for deviations of β for $4s$ photoelectrons of scandium outside resonances. They point out, however, that the total cross section was too small to allow detection of β at those regions. We hope that our results will greatly stimulate further theoretical and experimental work.

In conclusion, we have reported measurements of the photoionization angular distribution parameter β using synchrotron radiation and laser cooled and trapped atoms. This novel technique allows precise determinations of β outside resonances and close to ionization thresholds. Substantial deviations of β from the nonrelativistic value of two observed for cesium $6s$ electrons outside autoionization resonances and below the $5p$ threshold could not be explained by a Cooper minimum, but are due to interchannel coupling [8], i.e, quantum interference among final state wave functions. Other atoms that can be laser cooled and are candidates for similar studies are the metal-alkaline, alkaline-earth, and metastable noble gases. Prospects include the study of photoionization of atoms in excited states, spin aligned samples [23], and ultracold molecules formed by photoassociation [24].

This work was supported by FAPESP, CNPq, CAPES, and CEPOF. Lending of optical components from LNLS, UNICAMP, USP-São Carlos, and CEPOF are acknowledged. We thank the excellent support from the LNLS staff, and D.Ugarte, A. Rubens Brito, F. Paixão Filho, L. E. Guimarães, and M. A. P. Lima for the critical reading of this manuscript.

*Present address: LNLS, Campinas, Brazil.
†Corresponding author.
Electronic address: arnaldo@lnls.br

[1] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One and Two-Electron Atoms* (Springer-Verlag, Berlin, 1958).
[2] V. Schmidt, *Electron Spectrometry of Atoms using Synchrotron Radiation* (Cambridge University Press, Cambridge, England, 1997).
[3] H. J. Metcalf and P. van der Straten, *Laser Cooling and Trapping* (Springer-Verlag, Berlin, 1999).
[4] D. S. Hall, Am. J. Phys. **71**, 649 (2003).
[5] B. DeMarco and D. S. Jin, Science **285**, 1703 (1999).
[6] See, for example, J. Kinast *et al.*, Phys. Rev. Lett. **92**, 150402 (2004); M. Greiner *et al.*, Nature (London), **426** 537 (2003); **415**, 39 (2002).
[7] A. F. Starace, in *Atomic, Molecular, and Optical Physics Handbook*, edited by G. F.W. Drake (AIP, New York, 1996), Chap. 24.
[8] Z. Altun and S. T. Manson, Phys. Rev. A **61**, 030702 (2000).
[9] X. Flechard *et al.*, Phys. Rev. Lett. **87**, 123203 (2001).
[10] S. T. Manson and A. F. Starace, Rev. Mod. Phys. **54**, 389 (1982).
[11] L. H. Coutinho *et al.*, Rev. Sci. Instrum. (to be published).
[12] V. L. Jacobs, J. Phys. B **5**, 2257 (1972).
[13] S. B. Whitfield *et al.*, Phys. Rev. A **66**, 060701(R) (2002).
[14] J.W. Turkstra *et al.*, Phys. Rev. Lett. **87**, 123202 (2001), and references therein.
[15] V. Mergel *et al.*, Phys. Rev. Lett. **74**, 2200 (1995).
[16] S. Wolf and H. Helm, Phys. Rev. A **56**, 4385 (1997); **62**, 043408 (2000).
[17] O. Hemmers *et al.*, Rev. Sci. Instrum. **69**, 3809 (1998); Phys. Rev. Lett. **91**, 053002 (2003).
[18] Th. Prescher *et al.*, J. Phys. B **19**, 1645 (1986).
[19] That is, recoil energy, source size, degree of linear polarization, thermal Doppler contribution, instrumental resolution, cross sections, and asymmetry parameters.
[20] J.-J. Yeh, *Atomic Calculation of Photoionization Cross-Sections and Asymmetry Parameters* (Gordon and Breach, New York, 1993).
[21] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California, Berkeley, 1981).
[22] S. B. Whitfield *et al.*, Phys. Rev. A **66**, 060701 (2002).
[23] M. Zielonkowski *et al.*, Europhys. Lett. **44**, 700 (1998).
[24] J. Weiner *et al.*, Rev. Mod. Phys. **71**, 1 (1999).
[25] At this energy we are above the $5s$ threshold (28.2 eV) as well, but the ionization cross section of this level is negligible compared to the $5p$.