Circular dichroism in the polarization of the fluorescence resulting from the decay of photoionized Ca atoms

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Circular dichroism has been detected in the polarization of the fluorescence resulting from the decay of excited calcium ions. Synchrotron radiation was used to both ionize and excite calcium atoms from their ground state, and the photoejected electron was detected in coincidence with the polarized fluorescent photon. By introducing a degree of left, then right circularly polarized light into the incident photon beam the circular dichroism could be measured. The analysis of the results was carried out using recent developments of the statistical tensor method, and it was possible to determine the sign of the phase difference between the outgoing waves.

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INTRODUCTION

It is well known that circular dichroism, characterized by the different response to right and left circularly polarized photon beams, is manifested in the photoionization of polarized atoms (see, for example, [1–3]). Circular dichroism in photoelectron angular distributions has also been predicted for direct double ionization of nonpolarized atoms when the two electrons are observed in coincidence [4,5], and in the case of Auger electron emission, provided the Auger electron is observed in coincidence with the photoelectron [6,7]. Similarly circular dichroism has been predicted for the photoionization process where the angular distribution or polarization of the fluorescence resulting from the decay of an excited ion is measured in coincidence with the photoelectron. A detailed theoretical analysis of the angular correlation between the photoelectron and subsequent polarized fluorescent photon is given elsewhere [8]. In this paper we report an observation of the circular dichroism that appears in the fluorescence polarization measured in coincidence with the photoelectron.

We have studied the photoionization of Ca atoms in a partial channel populating the $Ca^+4p\ ^2P_{3/2}$ level. The angle-resolved photoelectrons have been detected in coincidence with the polarized fluorescent photon arising from decay of the $Ca^+\ 4p\ ^2P_{3/2}$ level to the Ca^+ ground state $4s\ ^2S_{1/2}$. The partial cross section for this process is very small ($\leqslant$0.1 Mb), but strong enhancement of this cross section ($\geqslant$10 Mb) occurs at the $3p^64s^2\ ^1S_0 \rightarrow 3p^54s^23d\ ^1P_1$ resonance [9]. In our previous paper [10], we reported a coincidence measurement between the angle-resolved photoelectron and the polarization-analyzed fluorescent photon in this giant resonance region. We demonstrated that this coincidence experiment could be regarded as a new approach to the "complete" experiment, from which the ratio of the dipole amplitudes (in LS coupling) and the magnitude of their relative phase could be extracted. However, the sign of the relative phase remained undetermined, and there was a large uncertainty in the values of both the dipole moment ratio and the magnitude of the phase difference.

In the present paper, our observation of circular dichroism allows us to determine the sign of the relative phase. This is another important piece of information from the viewpoint of the "complete" experiment; only the absolute scale of the dipole amplitudes remains to be determined in the present case.

THEORETICAL BACKGROUND

We choose the coordinate frame so that the y axis is directed along the incident photon beam and the z axis is along the principal axis of its linear polarization (see Fig. 1). We consider the case in which the fluorescent photons are detected in the x-axis direction in coincidence with the electrons detected in the z-x plane. If the detector is insensitive to the fluorescent photon polarization, the twofold differential cross section for the photoionization of the atom that emits a photoelectron e and a fluorescent photon γ can be expressed as follows:

$$\frac{d^2\sigma}{d\Omega_e d\Omega_\gamma} = \sigma(\theta_e)\ \frac{\omega_\gamma}{4\pi}\{1-\tfrac{1}{2}\alpha_2^\gamma[\mathscr{A}_{20}(J;\theta_e) - \sqrt{6}\mathscr{A}_{22}(J;\theta_e)]\}. \quad (1)$$

In this equation we have used the density matrix and statistical tensor formalism [8]; the components of the normalized

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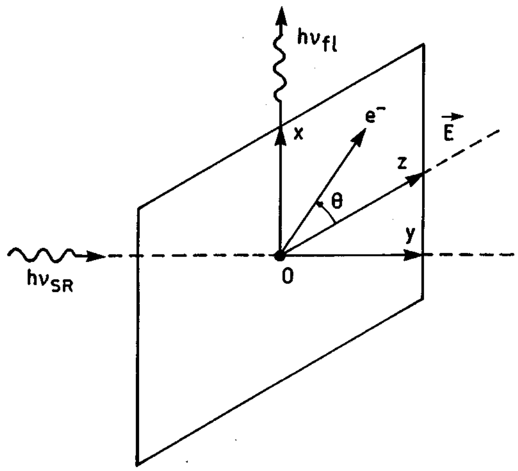

FIG. 1. Experimental geometry.

second-rank statistical tensor (alignment tensor) that describe the ionic state J produced by photoionization with the photoelectron detected in the direction θ_e are given by the equation

$$\mathscr{A}_{2q}=\rho_{2q}(J,J;\theta_e)/\rho_{00}(J,J;\theta_e). \tag{2}$$

In Eq. (1) $\sigma(\theta_e)$ corresponds to the differential photoionization cross section (with respect to θ_e), ω_γ is the fluorescence yield, and α_2^γ is the fluorescence decay anisotropy coefficient. For the particular transition $4p\ ^2P_{3/2}\rightarrow 4s\ ^2S_{1/2}$ considered here $J=\frac{3}{2}$, $\omega_\gamma=1$, and $\alpha_2^\gamma=\frac{1}{2}$. The statistical tensors $\rho_{2q}(J,J;\theta_e)$ can be expressed in terms of the statistical tensors of the incident photon beam (Stokes parameters) and the photoionization amplitudes and their relative phases. The explicit expressions are given by Kabachnik and Ueda [8].

The cross section of Eq. (1) can be presented in the form

$$\frac{d^2\sigma}{d\Omega_e d\Omega_\gamma}=A_0+A_2\cos 2\theta_e+A_4\cos 4\theta_e+B_2\sin 2\theta_e\equiv I(\theta_e), \tag{3}$$

where A_0, A_2, and A_4 depend on the degree of linear polarization S_1, whereas B_2 is proportional to the degree of circular polarization S_3. Circular dichroism in the angular distribution of the photoelectrons, which is exhibited in the difference between the intensities of photoelectrons ejected by right and left circularly polarized photons, can thus be expressed as

$$I(\theta_e,S_3=1)-I(\theta_e,S_2=-1)=2B_2\sin 2\theta_e. \tag{4}$$

If LS coupling is valid, then only the s and d photoelectron partial waves contribute to the photoionization in this channel, and B_2 is proportional to $-|D_s||D_d|\sin(\delta_s-\delta_d)$. Here we define the phase δ assuming positive amplitudes of the dipole moments $\mathscr{D}_i=|D_i|\exp(i\delta_i)$. It is thus clear that the sign of the relative phase $\Delta=\delta_S-\delta_d$ can be determined from the observation of circular dichroism. Note also, following Eq. (4), that the circular dichroism disappears at $\theta_e=n\pi/2$, n integer.

Alternatively, if the degree of linear polarization of the fluorescent photons, $P_x(\theta_e)=(Iz-I_y)/(I_z+I_y)$, with I_z and I_y being the yields of fluorescent photons polarized along the z and y axes, respectively, is measured in coincidence with photoelectrons detected at an angle θ_e, $P_x(\theta_e)$ can be expressed as

$$P_x(\theta_e)=\frac{\alpha_2^\gamma[3\mathscr{A}_{20}(J;\theta_e)+\sqrt{6}\mathscr{A}_{22}(J;\theta_e)]}{\alpha_2^\gamma[\mathscr{A}_{20}(J;\theta_e)-\sqrt{6}\mathscr{A}_{22}(J;\theta_e)]-2}. \tag{5}$$

$P_x(\theta_e)$ can also reveal circular dichroism; such dichroism, defined by the expression $P_x(\theta_e,S_3=1)-P_x(\theta_e,S_3=-1)$ is similarly proportional to $-|D_s||D_d|\sin(\delta_s-\delta_d)\sin 2\theta_e$ if the dichroism is small.

EXPERIMENT

The experimental apparatus and procedure are similar to those reported previously [10]. Briefly, the experiment was carried out on the toroidal grating monochromator fitted to the atomic and molecular science beam line at the Daresbury Synchrotron Radiation Source. At a photon energy of approximately 30 eV the monochromator has a resolution of ~50 meV and a peak flux ~10^{11} photons/sec. The degree of linear polarization S_1 of the incident light was 0.78 ± 0.02, determined from previous measurements, and the circular polarization parameter S_3 was unknown within the range $|S_3|\leqslant 0.6$. The electron spectrometer was a 90° spherical sector electrostatic analyzer of 90-mm mean orbit radius and was fixed at an angle of $\theta_e=135°$ (see Fig. 1 for the definition of θ_e). Photoelectrons leaving the Ca^+ ion in the $4p$ excited state were detected without resolving the spin-orbit components $^2P_{1/2}$ and $^2P_{3/2}$. Fluorescent photons arising from the subsequent $4p\ ^2P_{3/2}\rightarrow 4s\ ^2S_{1/2}$ transition in Ca^+ passed through a polarizer before being detected in the direction of the x axis in Fig. 1. A filter was used to remove the unpolarized $4p\ ^2P_{1/2}\rightarrow 4s\ ^2S_{1/2}$ component. The coincidence measurement of fluorescence polarization P_x was carried out at the peak of the $3p^64s^2\ ^1S_0\rightarrow 3p^54s^23d\ ^1P_1$ resonance at 31.41 eV.

Three separate measurements were made: first, the whole beam-line aperture was used, i.e., equal amounts of light above and below the storage orbit plane were accepted, as is normal practice for this beam line, and a coincidence polarization measurement was made. Second, by using the adjustable aperture in front of the diffraction grating, light falling onto the upper part of the grating was masked off, and the polarization measured. A third polarization measurement was made with the lower part of the grating masked off. For these half aperture measurements, the independent movable baffles that form the grating mask were adjusted so as to exactly halve the photon intensity measured at the experiment. In this way, and bearing in mind any reflection inversions caused by the beam-line optics, we introduced a degree of either left (upper part of grating obscured) or right (lower part obscured) circularly polarized light into our experiment. Our definition of left and right is made with the observer looking in the same direction as the incident beam is traveling.

RESULTS AND DISCUSSION

In our previous measurements of P_x at $\theta_e=-90°$ we found P_x to be ~0.6 at the peak of the $3p^64s^2\ ^1S_0$

$\rightarrow 3p^5 4s^2 3d\ ^1P_1$ resonance; this indicates that LS coupling is valid for the relevant photoionization channel at this resonance (see discussions in [8,10]). By taking a weighted mean of the three measurements of P_x at $\theta_e=-135°$ in the $3p$-$3d$ peak region [10], we obtain $P_x=0.459\pm0.021$ [10]. The ratio of the dipole amplitudes $|D_s|/|D_d|$ and their relative phase $|\Delta|$, which were determined in Ref. [10], were subject to a wide range of uncertainty owing to the unknown value of S_3 ($|S_3|\leqslant 0.6$). The present measurement of P_x at $\theta_e=135°$ using the entire area of the grating (first measurement to which we refer above) resulted in $P_x=0.512\pm0.045$. Although these two values of P_x at $\theta_e=\pm135°$ agree within the sum of their two uncertainties, they may be affected by contributions from the S_3 terms, whose signs are opposite for these two cases. Within the error of our measurements, we can, to first approximation, correct for any dependence on S_3 by taking the average of these two values; this gives a value of $P_x=0.477\pm0.034$. The values of $|D_s|/|D_d|$ and $|\Delta|$ obtained from this value of P_x are

$$|D_s|/|D_d|=1.6+0.4,-0.2, \quad |\Delta|=44.2°+2.0°,-2.5°. \tag{6}$$

We have calculated P_x as a function of θ_e using the present values of $|D_s|/|D_d|$ and $|\Delta|$ and assuming $S_1=0.78$. The polar plots of $P_x(\theta_e)$ are shown in Fig. 2, where the solid, dashed, and dotted lines correspond to the cases for $S_3=0$, $|S_3|=0.6$ with $S_3\sin\Delta>0$, and $|S_3|=0.6$ with $S_3\sin\Delta<0$, respectively. As can be seen in the figure, significant circular dichroism is predicted even for an incident light beam that has a relatively large degree of linear polarization ($S_1=0.78$). Figure 2 also shows that the circular dichroism is predicted to disappear at $\theta_e=n\pi/2$, where n is integer and that $P_x\sim0.6$ at $\theta_e=(2n+1)\pi/2$.

In Fig. 2, we show two data points, obtained using partially right (positive S_3) and left (negative S_3) circularly polarized light, respectively (second and third measurements above), and our original data point at $\theta_e=-135°$; the full aperture data point at $\theta_e=+135°$ is omitted for purposes of clarity. The values of P_x are 0.528 ± 0.060 for positive S_3 and 0.401 ± 0.074 for negative S_3. The mean value of these two is close to the mean obtained from the two measurements at $\theta_e=\pm135°$. The larger P_x for positive S_3 at $\theta_e=+135°$ indicates a positive sign of $\sin\Delta$, as seen in Fig. 2, and thus

$$\Delta=\delta_s-\delta_d=44.2°+2°,-2.5°. \tag{7}$$

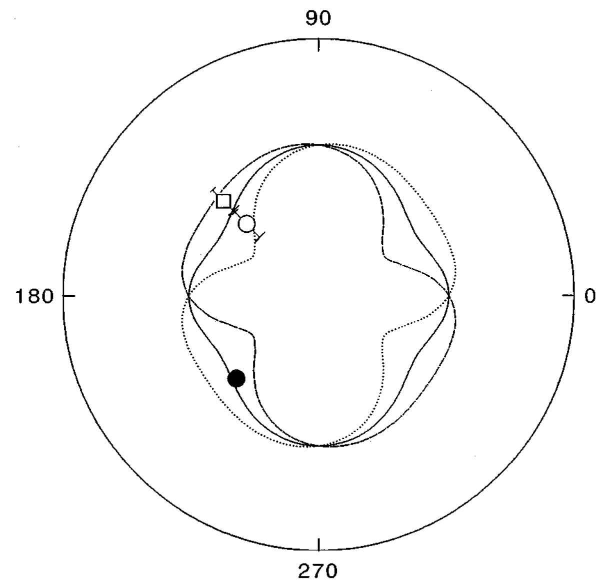

FIG. 2. Coincident fluorescence polarization angular map, where $P_x\sim0.6$ for $\theta=(2n+1)\pi/2,n$ integer. Calculated curves: for $\cdots$, $S_3\sin\Delta<0$ and ---, $S_3\sin\Delta>0$, $|S_3|=0.6$; —, $S_3=0$. ◇, measurement for S_3 positive and ○, for S_3 negative. ●, measurement from Ref. [10].

We emphasize that the measurements presented here are of sufficient accuracy to determine only the sign of $\sin\Delta$, and are not a measure of the degree of circular polarization contained in the photon beam incident on our experiment. They are, however, of sufficient accuracy to show that there is circular dichroism in the coincident fluorescence polarization for an unpolarized atomic target, and this has enabled us to determine the sign of the relative phase of the photoionization amplitudes. This provides similar information to angular correlation measurements between Auger electrons and photoelectrons using circularly polarized light [6,7,11].

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