New Approach for a Perfect Experiment: 2p Photoionization in Atomic Magnesium

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A new experimental approach for determining photoionization matrix elements which avoids the spin analysis of the photoelectron is presented for the case of 2p photoionization of atomic magnesium by 80-eV photons. Our experimental results clearly demonstrate the inadequancy of presently available theoretical calculations for this system.

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The analysis of a complete set of experimental data from which absolute values of the transition matrix elements and their relative phases can be extracted has been referred to as providing a *complete* (or perfect) description of the photoionization process.[1,2] The standard procedure for the acheivement of a complete description of a photoionization event has been to measure the absolute photoionization cross section and the angular distribution and spin polarization of the photoelectrons.[1,3,4] However, the low efficiency of Mott detectors used for the determination of the spin of the photoelectrons makes this measurement tedious and difficult. In this Letter we present an example (the 2p photoionization of magnesium) of an alternative experimental approach for the complete description of photoionization which is applicable when the ion is left in an aligned state.[5-8] The difficulties attendant to the spin-polarization measurement of the photoelectron is obviated by investigation of the decay of the aligned state of the ion which, of course, contains relevant information about the process.[9-12] Under appropriate conditions, the aligned state of the ion can be measured—if we assume a two-step model—by investigation of the nonisotropic angular distribution of the subsequent Auger electrons or by the nonisotropic angular distribution or polarization of the subsequent fluorescence radiation.[13-16] Previously, these decay modes were investigated separately: (i) Auger transitions[17,18]; (ii) polarization of fluorescence radiation[8,19,20]; (iii) angular distribution of fluorescence radiation.[21] However, in cases where the photoionization process is adequately described by two contributing matrix elements and the ion undergoes a simple radiative or nonradiative decay, it is possible to obtain complete information about the photoionization process by measurement of (i) the cross section, (ii) the angular distribution of the photoelectrons, and (iii) the alignment of the ion.

The 2p photoionization in atomic magnesium with its subsequent Auger decay is well suited for a demonstration of the proposed approach for complete information on a photoionization process. Of course, as discussed by Kessler,[2] any experiment can be considered complete (or perfect) only within the framework of a theory. For the 2p photoionization of magnesium, the Russell-Saunders coupling and the dipole approximation are applicable. Within this framework, the following relevant quantities are given in atomic units[22-24]: partial 2p photoionization cross section,

$$\sigma_{2p}=3\pi\{|D_s|^2+|D_d|^2\};$$

angular distribution parameter of 2p photoelectrons,

$$\beta_{2p}=\frac{|D_d|^2-\sqrt{8}|D_s||D_d|\cos\Delta}{|D_s|^2+|D_d|^2};$$

angular distribution parameter for the two fine-structure components of the subsequent Auger transitions,

$$\beta(L_2\text{-}M_1M_1)=0,$$

$$\beta(L_3\text{-}M_1M_1)=\frac{|D_s|^2+|D_d|^2/10}{|D_s|^2+|D_d|^2}.$$

The prerequisite for a nonisotropic angular distribution of any Auger transition is a nonvanishing alignment tensor[14,16,24,25] A_{20} of the photoion. This tensor is a measure of a nonuniform population of the magnetic sublevels of the photoionized state. Generally, the Auger angular distribution parameter is proportional to the alignment tensor.[12] For the present case, one simply gets

$$A_{20}(\ldots 2p^5 3s^2\,{}^2P_{3/2})=-\beta(L_3\text{-}M_1M_1).$$

In the above expressions, D_s and D_d represent the two dipole matrix elements. If all kinds of electron correlation effects are included, these matrix elements are complex quantities. In the limiting case of uncorrelated motion of the electrons, they are related to the ϵs and ϵd partial waves of the continuum electron and their relative phase reduces to the Coulomb- and short-range-potential phase differences of the partial waves.

The experiment is based on the method of angle-resolved electron spectrometry[26] and was performed at the Berlin electron storage ring BESSY at the toroidal grating monochromator (TGM4) with use of a photon energy of 80 eV. In our analysis we use the total absorption cross section at 80 eV given by Henke *et al.*[27] It should be noted that the tabulated total absorption data are based on transmission experiments on solid samples;

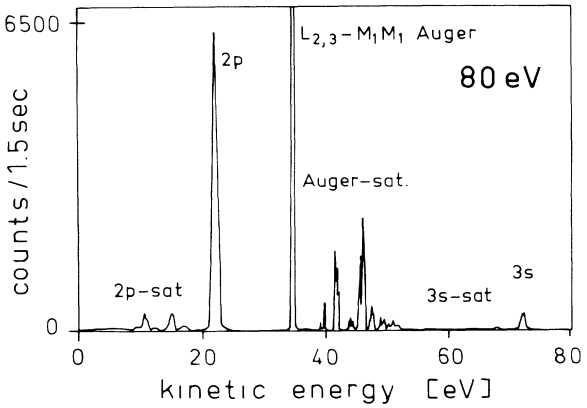

FIG. 1. Spectrum of electrons ejected from magnesium atoms after interaction with 80-eV photons. The spectrum was taken at the quasimagic angle of the electron analyzer (Ref. 26) in order to allow the extraction of relative intensities.

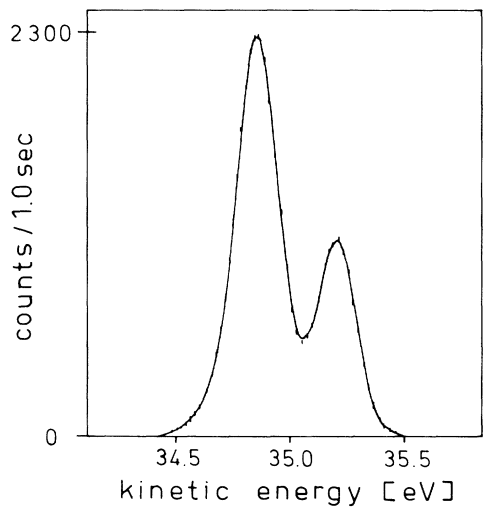

FIG. 2. Doublet of L_3-M_1M_1 and L_2-M_1M_1 Auger electrons taken at an angle of 33.3° with respect to the main axis of the polarization ellipse of the photon beam. Experimental data: points with error bars; resulting fitted curve: solid line.

however, they are expected[27] to represent atomic photoabsorption cross sections for photon energies above 50 eV and away from threshold regions. These conditions are fulfilled for the present case of $2p$ shell ionization. For photon energies around 80 eV, theoretical calculations[28,29] predict that $2s$ excitations with subsequent autoionizing decay which might influence the $2p$ photoionization process. This possibility was explored by measurement of the relative intensities of the $L_{2,3}$-M_1M_1 Auger lines and the $2p$ photoline around 80 eV. No peculiarity was found and we conclude that the $2s$ excitation does not influence the $2p$ photoionization process.

Figure 1 shows a complete spectrum of electrons ejected from magnesium atoms after photoionization at 80 eV. After subtraction of a constant background, application of a small correction for a smooth decrease of the analyzer transmission and detector efficiency at low kinetic energy,[30] and correction for the dispersion of the analyzer, relative intensities for individual photoprocesses were extracted from this spectrum. By use of the value for the total absorption cross section at 80 eV, $\sigma_{2p}=6.0$ Mb,[27] with an error of 0.5 Mb ascribed by us, the relative values were placed on an absolute scale. One then gets, at 80-eV photon energy, the following partial cross sections: $\sigma_{2p}=4.46(40)$ Mb, $\sigma_{3s}=0.080(11)$ Mb, σ_{2p}(discrete and continuous satellites) $=1.45(16)$ Mb, σ_{3s}(discrete and continuous satellites) $=0.014(4)$ Mb. The σ_{2p} is of special interest to the present work.

Because of the fine-structure splitting in the $L_{2,3}$ shell, the subsequent Auger transitions consist of two lines, L_2-M_1M_1 and L_3-M_1M_1. This doublet is shown in Fig. 2 together with the result of a standard least-squares fitting procedure based on the experimental line shape and the known energy separation.[31] Although both Auger components are not resolved completely in the present experiment, the excellent agreement between the fitted curve (solid line) and the experimental points demonstrates that intensities can be associated to each line with a high accuracy.

In order to determine the angular distribution of the $2p$ photoelectrons and the $L_{2,3}$-M_1M_1 Auger electrons, helium was also introduced into the target region for a simultaneous measurement of the polarization character of the monochromatized light.[32] In Fig. 3 the angle-dependent intensities are plotted for the $2p$ photoelectrons and the L_2-M_1M_1 and L_3-M_1M_1 Auger-electron components. The corresponding β parameters are $\beta_{2p}=0.74(2)$, $\beta(L_3$-$M_1M_1)=0.16(1)$, and $\beta(L_2$-$M_1M_1)=0.00(1)$. It is noteworthy that the experimental value of the last β parameter gives zero, which is in perfect agreement with the theoretical prediction that this line must be isotropic.[24]

Based on the experimental values for σ_{2p}, β_{2p}, and $\beta(L_3$-$M_1M_1)$ and on the theoretical expressions given above for these quantities with respect to the dipole matrix elements, one can calculate $|D_s|$, $|D_d|$, and Δ. Table I shows a compilation of our experimental values together with the results of several theoretical calculations which take into account different amounts of electron correlation. The calculations marked with HS and HF(^1P) are based on the model of uncorrelated motion of the electrons. HF(^1P) is expected to be the better value because the continuum function was calculated in a state-dependent Hartree-Fock potential which takes into account intrachannel electron correlations.[22] Generally, the RRPA calculation should be even better, since it accounts for many of the important aspects of electron correlations and for the relativistic interactions responsible for the spin-orbit effects. The comparison between the theoretical data and the experimental values shows that the relative phase Δ agrees sufficiently well for all cases, whereas the theoretical values for $|D_d|$ and the

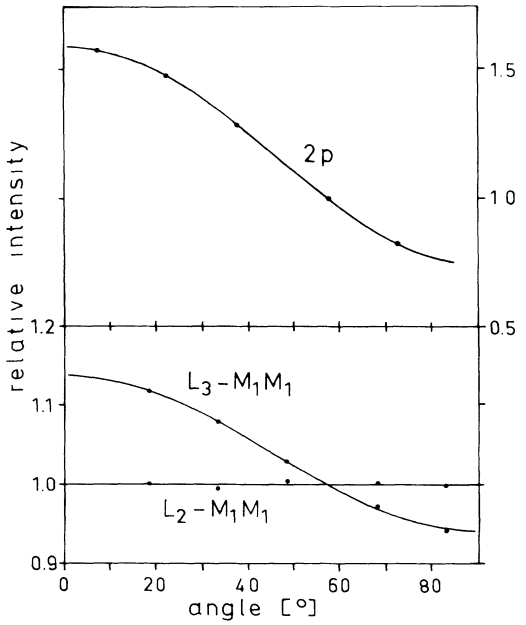

FIG. 3. Angle-dependent intensities: upper part for $2p$ photoelectrons, lower part for L_3-M_1M_1 and L_2-M_1M_1 Auger electrons. Experimental data: points; fitted curve for the determination of the β parameter: solid line. The angle is measured with respect to the main axis of the polarization ellipse of the light; the relative intensities were normalized to 1 at the quasimagic angle (different for the measurements of photoelectrons and Auger electrons, respectively, because of different runs).

TABLE I. Compilation of dipole matrix elements, absolute values and relative phase, for $2p$ photoionization in magnesium at 80-eV photon energy. Theoretical values are from relativistic random-phase approximation (RRPA, Ref. 33) and calculations based on a state-dependent Hartree-Fock potential [HF(^1P), Ref. 34] or on the Herman-Skillman potential (HS, Refs. 35–41).

	$\lvert D_s \rvert$ (a.u.)	$\lvert D_d \rvert$ (a.u.)	Δ (rad)[b]
Expt.	0.034(3)	0.126(6)	4.99(16)[c]
RPPA[a]	0.044	0.149	5.03
HF(^1P)[a]	0.038	0.147	4.82
HS	0.037	0.157	4.92

[a]These values were extracted from the figures of Ref. 33 at 84.4-eV photon energy in order to compensate for the difference in theoretical and experimental ionization thresholds.

[b]The phase difference Δ contains two contributions, the Coulomb phase difference σ and the phase difference δ caused by the short-range potential. σ can be calculated analytically in this case to be $\sigma=\sigma(\epsilon s)-\sigma(\epsilon d)=1.03$ rad which yields $\delta=\Delta-1.03$ rad.

[c]The experimental phase was extracted from $\cos\Delta$ only. In principle, this still leaves an ambiguity of the phase differences (Ref. 2); in the present case $\Delta=1.29(16)$ would also be a solution. However, the latter value is excluded by theory.

RRPA value for $|D_s|$ are significantly larger than experiment. These differences reflect a shortcoming of the treatment of electron correlations in the given theoretical approaches.

The general agreement between the values of all three calculations suggests that neither relativistic effects nor interchannel coupling, which are essential constituents of RRPA, play a major role in the case of $2p$ photoionization in magnesium. Improved calculations which are beyond RRPA have to account in a balanced way for the multiconfigurational nature of the ground and final ionic states (including core relaxation).[33,42–45] The importance of such correlation effects manifests itself also in the remarkable intensity of $2p$ satellite transitions found at 80-eV photon energy: σ_{2p}(all satellites)/σ_{2p}(main transition) $=0.33$. The sum of both contributions, i.e., $\{\sigma_{2p}+\sigma_{2p}$(all satellites)$\}$ yields 5.9(4) Mb, which agrees very well with the result of 6.1 or 6.4 Mb obtained in the HF(^1P) or RRPA calculation, respectively. Such a behavior was recognized already for the case of xenon $5s$ photoionization.[44] An intensity borrowing mechanism might be responsible for a redistribution of the oscillator strength from the single-particle calculation for $2p$ photoionization into the $2p$ main line and the $2p$ satellites. An adequate theory, therefore, should simultaneously provide for the following facts: a considerable reduction for the values of the single-particle $|D_d|$ and possibly the $|D_s|$ matrix elements, bringing them closer to the experimental values; an explanation for the insensibility of the relative phase Δ against electron correlation; the remarkable intensity of the $2p$ satellites; and agreement between theoretical and experimental ionization thresholds (cf. Refs. 33 and 46).

In conclusion, the experiment presented in this Letter describes an alternative way to achieve complete information about certain systems which avoids the generally difficult spin-polarization measurement. Experimental data of high quality are easily available by this method. These data provide a guide for further theoretical developments which consider the multiconfigurational nature of the ground and final ionic state of the atom.

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