INFLUENCE OF NUCLEAR SPIN ON ANGULAR DISTRIBUTION AND POLARIZATION OF PHOTOELECTRONS: RESONANT TWO-PHOTON IONIZATION OF Na

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Resonant ionization of Na through its 3p levels by two pulsed lasers illustrates how coherent excitation of hyperfine levels affects the angular distributions of photoelectrons and of electron spin polarization. The effects appear as dependence of the distributions on the time interval between excitation and ionization pulses. Theoretical and experimental results are given for photoelectron distributions, and several microscopic parameters are determined. Theoretical predictions are given for angle- and time-dependence of spin polarization.

1. Introduction

This report describes the influence of coherence in an excited state on angular properties of photoelectrons. In particular, we examine — both theoretically and experimentally — the effect of coherence among hyperfine states on the angular distribution of photoelectrons from the resonant two-photon process $Na(3s\,^2S) \rightarrow Na(3p\,^2P_{3/2}) \rightarrow Na^+ + e$, as excited by two linearly-polarized pulsed laser beams, and, as a theoretical prediction only, the angular dependence of electron spin polarization from ionization through $Na(^2P_{1/2}) \rightarrow Na^+ + e$ by two circularly-polarized beams. The effects are observed by variation of the time interval between the laser pulses.

Atomic sodium excited to its $^2P_{3/2}$ level with the 4 ns pulse of a dye laser tuned to 588.99 nm, enters a superposition of the hyperfine states of this level. A second light pulse, from a N_2-laser (337.1 nm), photoionizes the excited atoms. The theory outlined below shows that this angular distribution depends on the degree of coherence among the hyperfine levels, and on the time interval between the exciting pulse and the ionizing pulse. The distribution also depends on the shapes and degrees of coherence of the light pulses. Our observations imply that in our experiments, an essentially completely coherent superposition of intermediate hyperfine states is produced. The angular distributions vary with the time interval expected for complete or nearly complete coherence, according to the phenomenology of the theory. Moreover, one can extract from the angular distribution two microscopic parameters: the ratio of the radial matrix elements for the transition from the intermediate 3p level to the two final channels and the difference in the short-range phase shifts of the two final partial waves at the photoelectron energy of 0.64 eV. The theory shows, lastly, that the intensity of polarized electrons and their angular dependence are sensitive to the hyperfine coupling.

The experiments, carried out with apparatus modified from our previous system [1], involve two nearly-collinear beams of linearly-polarized light, with angle η between their polarization vectors, intersecting a beam of Na atoms at 90°. Photoelectrons in the ("equatorial") plane perpendicular to the propagation axis of the light beams are collected by a channeltron multiplier subtending a solid angle of about 7°. The angle between the polarization vector of the ionizing light of the N_2 laser and the momentum vector k of the electron is θ.

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2. Angular distribution of electrons

A phenomenological analysis of this system [2,3] gives the angular distribution of photoelectrons in the equatorial plane in terms of the intensity $\mathcal{I}(\theta)$, in units of the square of the initial → intermediate transition dipole *:

$$\mathcal{I}(\theta)/|\langle 3p|r|3s\rangle|^2 = C_{00}P_{00} + C_{20}P_{20}(\cos\theta) + C_{40}P_{40}(\cos\theta) + C_{21}P_{21}(\cos\theta) + C_{41}P_{41}(\cos\theta). \quad (1)$$

We now introduce microscopic theory; let $|s|$ and $|d|$ represent the absolute values of the radial one-electron matrix elements $\langle ks|r|3p\rangle$ and $\langle kd|r|3p\rangle$, respectively, δ_0 and δ_2 be the short-range contributions to the outgoing s-wave and d-wave phase shifts [4], and $\delta = \delta_0 - \delta_2$. Then the coefficients of (1) can be written

$$C_{00} = \tfrac{1}{27}[2\chi(0) + (2 - 3\sin^2\eta)\chi(2)]\,|s|^2 + [4\chi(0) + \tfrac{2}{5}(1 + 3\sin^2\eta)\chi(2)]\,|d|^2,$$

$$C_{20} = \tfrac{2}{27}(\tfrac{1}{5})^{1/2}[-2\chi(0) + (3\sin^2\eta - 2)\chi(2)] \times |s|\,|d|\cos\delta + [2\chi(0) + \tfrac{1}{7}(5 - 3\sin^2\eta)\chi(2)]\,|d|^2,$$

$$C_{40} = \tfrac{8}{105}(1 - 2\sin^2\eta)\chi(2)|d|^2,$$

$$C_{21} = \tfrac{1}{3}(\tfrac{2}{15})^{1/2}[-|s||d|\cos\delta + \tfrac{2}{7}|d|^2]\sin(2\eta)\chi(2),$$

and

$$C_{41} = \tfrac{4}{21}(\tfrac{1}{5})^{1/2}\sin(2\eta)\chi(2)|d|^2. \quad (2)$$

The factors $\chi(0)$ and $\chi(2)$ contain all information concerning the pulse shapes and the dynamics and coherence of the intermediate state. The intermediate state itself is generated by the dipole radiation of the first beam, with intensity $I_1(t)$, acting on an isotropic ensemble. Hence the density matrix of the intermediate state contains a monopole part $\rho_i^{(0)}(t)$ and a quadrupole part $\rho_i^{(2)}(t)$. The second light beam, with intensity $I_2(t)$, probes these two parts. The pulse shapes are convoluted with a field-independent factor to give a useful function [3] (see below):

$$\chi(J) = \int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} dt''\, I_1(t')I_2(t'')\,W_J(t'' - t'). \quad (3)$$

If $I_2(t'')$ were a δ-function, $\chi(J)$ would be the 2^J-pole polarization of the intermediate state at the instant of ionization.

$W_J(t'' - t')$ gives the time evolution, through the interval $\tau = t'' - t'$, of those elements of the density matrix of the intermediate level describing its electronic 2^J-pole polarization, with the (unobserved) nuclear spin unpolarized. W_J depends on the nuclear spin $i(3/2)$, the spin-plus-orbital angular momentum j, the total angular momenta f', the energies $E(f') = \hbar\omega'$ of the hyperfine states and the radiative lifetime Γ^{-1} of the intermediate level.

$$W_J(\tau) = \sum_{f',f''} \langle (ij)f'(ij)f''; J|(ii)0\,(jj)J; J\rangle^2 \times \exp[i(\omega' - \omega'')\tau]\exp(-\Gamma\tau). \quad (4)$$

The sum is over all pairs of final states with $|i - j| \leqslant |f| \leqslant |i + j|$. The bracket factor is the (real) recoupling coefficient for generating angular momentum j from i, j, f' and f'' taken in the orders indicated by the parentheses [5]. In physical terms, the coupling represented by the ket constructs J from spin and orbital angular momentum only, whereas the bra is constructed from stationary states of the hamiltonian including hyperfine interaction.

Typical measured angular distributions are shown in fig. 1, with curves of $\mathcal{I}(\theta)$ fit to eq. (1) by least squares. Absolute $\mathcal{I}(\theta)$'s were not measured so only ratio's of the C_{lm}'s were determined. Nonetheless, from (2) and measurements of $\mathcal{I}(\theta)$ as a function of η and τ, one can determine $r = |s|/|d|$, $\cos\delta$, and $\chi(2)/\chi(0)$.

The atomic parameters were derived by two methods: (1) directly from the C_{lm}'s obtained by fitting curves, as in fig. 1, for several values of η and with τ fixed (at 4 ns); (2) from two relations independent of η and τ, the ratio C_{21}/C_{41} and

$$\frac{\mathcal{I}(0)}{\mathcal{I}(\pi/2)} = \frac{(r^2 - 4r\cos\delta + 4)}{(r^2 + 2r\cos\delta + 1)}.$$

These were computed for all measured distributions

* Eq. (1) is essentially contained in ref. [2].

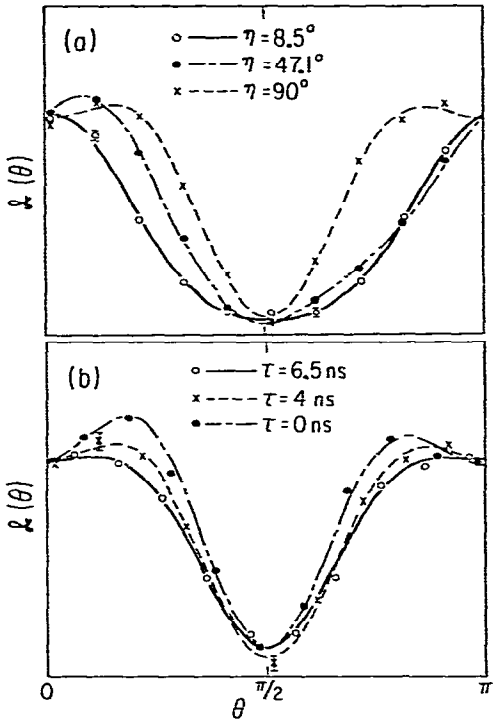

Fig. 1. Experimental (points) and theoretical (curves) angular distributions of photoelectrons: (a) with a constant 4 ns interval τ between the peaks of light pulses, but with different angles η between the polarization vectors of the two light beams; (b) with η fixed at 90 ± 3° but with different time intervals.

and their means were used to determine r and cos δ, from which $T_{20} = \chi(2)/\chi(0)$ was then found for each τ.

The best-fit atomic parameters are $r = 1.11 \pm 0.10$ and cos $\delta = 0.81 \pm 0.05$. The value of cos δ can be inferred from the extrapolated quantum defects $\eta_l = \delta_l - \arg\Gamma(l + 1 - ik^{-1})$ of Risberg [6] or from theoretical predictions. From quantum defect theory [4,7,8] we have $r = -0.646$ and cos δ between 0.88 and 0.92. If we use the experimental total cross section of Rothe [9], we obtain amplitudes (s and d) of −1.03 and 0.93, respectively, as compared with Seaton's [7] values of −0.643 and 0.996.

The angular distribution of photoelectrons is indeed a function of time; the higher harmonics of $\mathcal{I}(\theta)$ disappear in about 10^{-8} s as the interference among the hyperfine levels changes with the advancing phase of each state. Alternatively, one can look upon the time

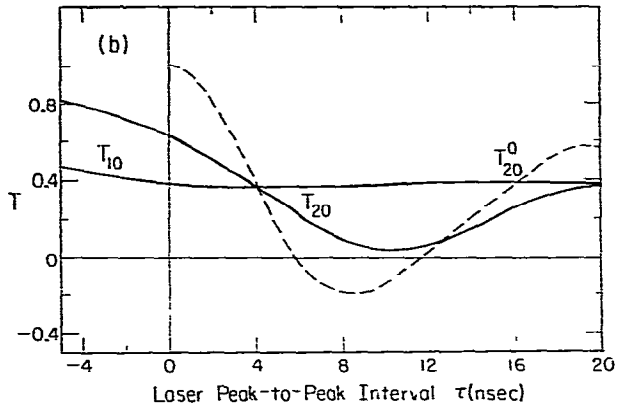

Fig. 2. The characteristics T_{20} and T^0_{20} for the $^2P_{3/2}$ level of sodium. T^0_{20} refers to excitation and ionization by δ-function pulses and T_{20} to the measured pulse shapes of our experiment. T_{10} for the $^2P_{1/2}$ level is also shown here.

dependence as a transfer of angular momentum polarization from the active electron to the nuclear spin.

The values of T_{20} from the experiments are 0.839 ± 0.060 for coincident pulses, 0.685 ± 0.030 for a delay of 4 ns and 0.414 ± 0.075 for a delay of 6.5 ns. If one assumes the radiation produces a coherent superposition of hyperfine levels, and that an integration over only the two times t' and t'' suffices in eq. (2) (rather than the rigorous convolution of four times, which would be necessary if the coherence time of the laser were 10^{-9} s or longer), and the shapes of the intensities $I_1(t), I_2(t)$ are known, then T_{20} (fig. 2) can be computed. If we assume the pulses are gaussian, the essential integrals can be done analytically and reduce to complex error functions. With these assumptions, which are only working hypotheses, we compute $T_{20} = 0.62$, 0.37 and 0.18 for the delays of 0, 4 and 6.5 ns, respectively. The discrepancies probably arise because the computed T_{20} values are based on the shaky assumption that I_2 is always so weak that ionization produces negligible depletion of the intermediate state. Hence the actual population of $^2P_{3/2}$ will always be younger and more polarized that that assumed in the simple theoretical model.

3. Angular distribution of spin polarization

The analysis can be extended to predict the effects

of hyperfine interactions on electron spin polarization produced by two collinear, circularly-polarized laser beams. The principal new consideration here is the dipole polarization in the intermediate state, which generates a term in $\chi(1)$.

Electron spin polarization is attainable from alkali atoms by resonant two-photon ionization with circularly-polarized radiation [10–12]. With collinear light beams, the electrons emerging at 90° to the propagation axis would all have β-spins, and most of the electrons would emerge in approximately this ring. Prior theoretical analyses neglected the influence of hyperfine coupling, but Granneman et al. [11] showed that it sometimes affects the total polarization drastically. We give results here for the angular distribution of spin-polarized electrons for the process $Na(^2S_{1/2}) \rightarrow Na(^2P_{1/2}) \rightarrow Na^+(^1S_0) + e(ks, kp)$. The more complex process through the $^2P_{3/2}$ intermediate level has also been analyzed and will be discussed in a fuller presentation.

Let $T = T_{10} = \chi(1)/\chi(0)$; no $\chi(2)$ appears for the $^2P_{1/2}$ state. The spin-independent average intensity $\mathcal{I}(\theta)$ for two-photon ionization by two circularly-polarized photons, through the $^2P_{1/2}$ state, is

$$\mathcal{I}(\theta; {}^2P_{1/2}) = A\{(\tfrac{2}{3}(1-T)(r^2+1-2r\cos\delta) + [(1+2T)+(1-T)2r\cos\delta]\sin^2\theta\}, \quad (5)$$

where now θ denotes the angle between the propagation axis of the electron and the propagation axes of the photons. The total cross section, obtained by integrating (5), corresponds to a rhs of $A[(\frac{2}{3}(1-T)r^2 + \frac{2}{3}(2+T)]$. With both light beams circularly polarized in the same direction, the intensity of electrons with α-spin is

$$\mathcal{I}^\alpha(\theta; {}^2P_{1/2}) = A\{\tfrac{2}{3}(1-T)(r^2+1-2r\cos\delta) + [(7T-1)/2+(1-T)2r\cos\delta]\sin^2\theta - 3T\sin^4\theta\}, \quad (6a)$$

and the intensity of electrons with β-spin is

$$\mathcal{I}^\beta(\theta; {}^2P_{1/2}) = A\{\tfrac{3}{2}(1-T)\sin^2\theta + 3T\sin^4\theta\}. \quad (6b)$$

(The axis of spin quantization is the propagation axis of the laser beams.) The intensity of polarized electrons as a function of θ is given by

$$\mathcal{I}^\alpha(\theta) - \mathcal{I}^\beta(\theta) = A\{\tfrac{2}{3}(1-T)(r^2+1-2r\cos\delta) + [(5T-2)+(1-T)2r\cos\delta]\sin^2\theta - 6T\sin^4\theta\}. \quad (7)$$

The function T, which depends on the interval between the arrival of the two photons, lies between the theoretical bounds of ±1. Hence the total intensity becomes $2A$ when $T = 1$. When the hyperfine levels are coherently excited, $T = 1$ and the ensemble is an isotropic; when $T = 0$, the electronic state of the ensemble is isotropic and the photoelectrons are unpolarized. Moreover, when $T = 1$, $\mathcal{I}^\alpha(\theta)$ and $\mathcal{I}^\beta(\theta)$ reduce to the form they have in the simple fine-structure analysis, so that the electrons emerging at $\theta = 90°$ will all have β-spin. However, the evolution of the hyperfine states causes an exchange of electron spin angular momentum polarization with the polarization of other angular momenta of the system so that some α-spin is introduced even at $\theta = 90°$, when $T < 1$.

For δ-function pulses $T = T_{10}^0$ is given by

$$T_{10}^0 = \tfrac{3}{8} + \tfrac{5}{8}\cos\omega_{21}t,$$

where $\omega_{21} = 1190$ MHz. The period of the oscillations is 5.3 ns. The effect of laser intensity functions similar to those for our N_2 and rhodamine lasers (fwhm = 8 ns and 4 ns, respectively) is to obscure completely the time variations in T_{10}. The resulting polarization

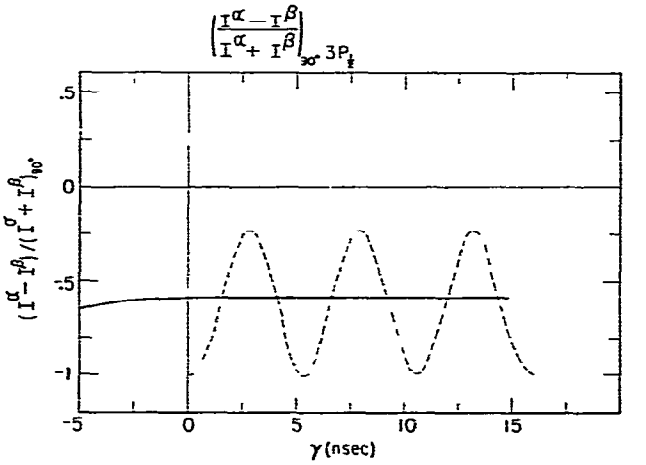

Fig. 3. Degree of polarization at $\theta = \pi/2$ for electrons produced by two collinear, circularly-polarized beams with the $^2P_{1/2}$ level as the intermediate state. The dashed curve represents δ-function pulses and the solid curve the measured pulse shapes of our experiment.

at $\theta = 90°$ for both situations is shown in fig. 3. If both laser pulses are brief, oscillations occur in the electron polarization which depend on the delay between pulses. If, however, extended laser pulses similar to ours are used, there is almost no observed variation in polarization with time; $(\mathcal{P}^{\alpha} - \mathcal{P}^{\beta})/(\mathcal{P}^{\alpha} + \mathcal{P}^{\beta})$ remains nearly fixed at −0.6. Laser pulses shorter than 1 ns arriving almost simultaneously would be necessary to produce completely polarized electrons by resonant two-photon ionization of sodium.

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