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Depolarization effects in pulsed photoionization of state-selected lithium[*]

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Depolarization effects in pulsed photoionization of state-selected lithium atoms have been observed. Experimental evidence and theoretical analysis demonstrate that these effects are produced by resonant excitation of the $2\,^2P$ state. Based upon the analysis, it is argued that resonant excitation of the $2\,^2P$ state with circularly polarized cw laser light followed by broad-band pulsed ionizing radiation can be used to produce an intense highly polarized electron beam with longitudinal polarization.

During the last few years interest in multiphonon processes has grown substantially. A number of theoretical studies[1-4] have specifically addressed the subject of the polarization of photoelectrons produced by multiphoton ionization of alkali-metal atoms. Recently, several experiments were reported in which the electron polarization was measured for the case of resonant two and three-photon ionization of sodium and cesium by circularly polarized light.[5,6]

In this paper we report on the observation of depolarization effects produced by resonant excitation in pulsed ionization of polarized lithium atoms by unpolarized light. Since the effect of the spin-orbit interaction in the continuum is negligible for lithium,[7] the depolarization is solely the result of the fine-structure coupling in the intermediate state. The role of the hyperfine interaction, it should be noted, is minimized, because the nuclear and electronic spins are effectively decoupled by an external magnetic field.

The measurements which we will describe were carried out on the polarized electron source at the Stanford Linear Accelerator Center (SLAC).[8,9] In this source, a beam of lithium atoms (95.6% Li^6 and 4.4% Li^7) is prepared in the $2\,^2S_{1/2}$ $m_j = +\frac{1}{2}$ state by high-field state selection in a six-pole magnet. Unpolarized broad-band light from a vortex-stabilized argon flash lamp is focused coaxially onto the lithium beam to produce a pulsed beam of highly polarized electrons with intensity $\sim 2\times 10^9$ e/pulse and polarization 0.85. In order to reduce the depolarizing effect of the hyperfine interaction, an axial field of 200 G is impressed upon the atoms in the ionization region. The design of the SLAC source is based upon a prototype which was the subject of earlier extensive investigation.[7] It was during those investigations that depolarizing effects in pulsed photoionization of alkali atoms were first observed,[10] but detailed studies were not pursued. In the early stages of development of the SLAC source the effects were not visible since the intensity of the resonance radiation was too low. However, as the light intensity was increased, the depolarization became prominent and detailed measurements were undertaken.

During the course of the measurements, two six-pole magnets were used, one with a 3.2-mm diameter bore and one with a 6.4-mm bore. For the small gap case, the electronic polarization, P_e^0, of the atoms in the ionization region is calculated to be 0.935 ± 0.050, the reduction below unity being attributable to residual hyperfine effects in the ionization region (5.5%) and to molecular contamination of the atomic beam ($\lesssim 1\%$). For the large gap case, where the high-field state selection is incomplete, P_e^0 is calculated to be 0.847 ± 0.050.

The electron polarization averaged over a pulse, $\bar{P}_e$, was determined experimentally by Mott scattering at 70 keV and Møller scattering at GeV energies. For the small gap case the Møller measurements indicated that the electron polarization was only 0.76 ± 0.03,[11] in considerable disagreement with the theoretical value. Mott measurements displayed a similar discrepancy. Studies of the photoelectron intensity as a function of the incident photon intensity, I_γ, suggested that the discrepancy between the predicted and experimental values of the electron polarization might be attributable to the presence of a multi-step photoionization process. As the data of Fig. 1 show, the electron yield per atom, I_e, contains a quadratic dependence on I_γ. The curve shown for I_e/I_γ is the best fit of the equation

$$I_e = I_\gamma + bI_\gamma^2 . \tag{1}$$

to the data, from which a value of $b = 0.535 \pm 0.072$ is obtained. The units of I_γ and I_e have been chosen to give $I_e/I_\gamma = 1$ at $I_\gamma = 0$.

Since the spectrum of the flash lamp does not have any prominent line structure, comparison of line strengths for transitions to various intermediate states indicates that the multistep photoionization process was probably proceeding via the $2S$-$2P$ resonant transition at 670.8 nm. Measurements with cut-off filters which restricted the bandwidth of the flash-lamp radiation verified this hypothesis; furthermore, when a broad-band uv interference filter was inserted to remove the 670.8-nm radiation, the quadratic term in Eq. (1) vanished and $\overline{P}_e$ was measured to be 0.850 ± 0.075 for the large gap magnet, in excellent agreement with the calculated value, P_e^0.

With the interference filter removed, the dependence of $\overline{P}_e$ on I_γ was measured by Møller scattering. The results of these measurements, normalized to P_e^0, are also shown in Fig. 1. We can develop a theoretical analysis of the excitation and ionization mechanism to explain the dependence of $\overline{P}_e/P_e^0$ on I_γ by first assuming that during the 1.6 μsec duration of the light pulse on atom may be excited to the $2P$ state one or more times before it is photoionized from either the $2S$ ground state or the $2P$ excited state. For the case of light incident parallel to the magnetic field, it can be shown using the appropriate line strengths and Clebsch-Gordan algebra that the depolarization factor associated with each excitation is 5/9. The photoelectron polarization thus depends upon the number of excitations executed and, as a consequence, upon time.

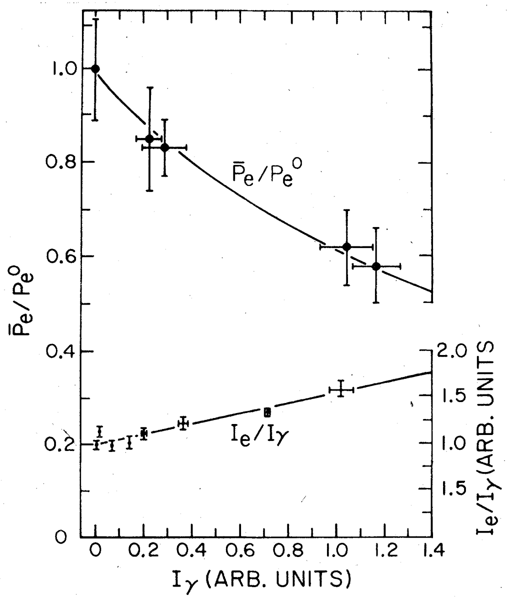

FIG. 1. Electron yield per atom, I_e, per unit light intensity, I_γ, and pulse average electron polarization, $\overline{P}_e$, per unit electronic atom polarization, P_e^0, as functions of I_γ.

We now define $N^S(t)$ and $N^P(t)$ as the number of atoms in the $2S$ and $2P$ states, respectively, at time t. We also define $N_\Delta^S(t)$ as the population difference of the two magnetic sublevels ($m_s = \pm\frac{1}{2}$) of the ground state and $N_\Delta^P(t)$ as the population difference of the two sublevels formed by projecting the $2P$ eigenstates onto $|m_s = \pm\frac{1}{2}\rangle$. Since the time structure produced by the fine-structure coupling is not resolved, we treat the $2P$ eigenstates as an incoherent superposition. Then with r denoting the $2P$ excitation rate, Γ the $2P$ spontaneous emission rate, and $R_S(R_P)$ the $2S(2P)$ ionization rate, we can write the following set of rate equations:

$$\dot{N}^S(t) = -(r + R_S)N^S + \Gamma N^P , \tag{2}$$

$$\dot{N}^P(t) = -(\Gamma + R_P)N^P + rN^S , \tag{3}$$

$$\dot{N}_\Delta^S(t) = -(r + R_S)N_\Delta^S + \Gamma N_\Delta^P , \tag{4}$$

$$\dot{N}_\Delta^P(t) = -(\Gamma + R_P)N_\Delta^P + \tfrac{5}{9}rN_\Delta^S , \tag{5}$$

where we have neglected stimulated emission[12] which was always <1.5% of the total emission for our light intensities. We define the instantaneous photoelectron polarization, $P_e(t)$, as

$$P_e(t) = \frac{R_S N_\Delta^S(t) + R_P N_\Delta^P(t)}{R_S N^S(t) + R_P N^P(t)} , \tag{6}$$

and obtain the average polarization $\overline{P}_e$ of all photoelectrons emitted during a pulse by integrating the numerator and denominator of Eq. (6) over t for the duration of the pulse.

Under our experimental conditions, the light intensity was sufficiently low that ≲3% of all atoms were photoionized and the average number of $2S$-$2P$ excitations per atom per pulse, α, was estimated to be of the order of one. Since the lifetime of the $2P$ state, $1/\Gamma$, is 27 nsec, the average time spent by an atom in the $2P$ state is negligible compared to the 1.6-μsec pulse duration, τ. Under these conditions we can obtain a relatively simple expression for $\overline{P}_e$. If we neglect the 3% depletion of atoms and assume that $\alpha \sim 1$, then for a square light pulse uniformly illuminating the atomic beam, we have $\alpha = r\tau$ and

$$\overline{P}_e = P_e^0 \frac{1 + 5\beta/9}{1 + \beta} \frac{1 - \exp(-4\alpha/9)}{4\alpha/9} , \tag{7}$$

where $\beta = bI_\gamma$ is the ratio of the $2P$ to $2S$ photoelectrons. Since α is proportional to I_γ we can rewrite Eq. (7) as

$$\overline{P}_e = P_e^0 \frac{1 + 5bI_\gamma/9}{1 + bI_\gamma} \frac{1 - \exp(-aI_\gamma)}{aI_\gamma} , \tag{8}$$

where $a = 4\alpha/9I_\gamma$. With $b = 0.535 \pm 0.072$ as determined from Eq. (1), the best fit of Eq. (8) to the

$\bar{P}_e/P_e^0$ data of Fig. 1 gives $a = 0.68 \pm 0.19$. For $I_\gamma^{\max} = 1.16 \pm 0.01$ we then have $\alpha^{\max} = 1.77 \pm 0.50$, consistent with our assumption that $\alpha \sim 1$. For a critically damped light pulse of the form $I_\gamma(t) = k(t/t_0)\exp(-t/t_0)$ with $t_0 = 0.5$ µsec and k chosen such that the pulse average value of $I_\gamma(t)$ is I_γ, a similar analysis gives $a = 0.71 \pm 0.20$. The fitted $\bar{P}_e/P_e^0$ curve for the critically damped light pulse is shown in Fig. 1.

The associated time dependence of the polarization implied by Eq. (6) was studied experimentally by either Mott or Møller scattering for three values of I_γ; namely, $I_\gamma = 0.23$, 1.04, and 1.61. Electron scattering events were recorded as a function of the elapsed time within the pulse, and each event was assigned to one of five time bins for the Mott measurement or one of four time bins for the Møller measurements. The results of the measurements are shown in Fig. 2. With a and b already determined and $\Gamma = 37 \times 10^6$ sec^{-1} known from spectroscopic measurements, only one of the four parameters used in Eqs. (2)–(5) is independent. Numerical integration of Eqs. (2)–(5) was thus used to obtain a one-parameter fit of $P_e(t)$ to the Mott data for $I_\gamma = 1.61$. The result of the fit with $r = 1.6$, $R_S = 0.026$ and $R_P = 0.48$ (all in units of 10^6 sec^{-1}) is shown by the solid curve in Fig. 2, which has a $\chi^2 = 3.9$ for 4 degrees of freedom. The dashed curves for $I_\gamma = 0.23$ and 1.04 were generated from the same values of r, R_S and R_P scaled according to I_γ. It should be noted that the values of R_S and R_P are consistent with the expected ratio $R_P/R_S \approx 20$, based upon the known $2S$ and $2P$ photoionization cross sections[13] and the measured lamp spectrum.

As a corollary to our studies, we make the observation that if the 670.8-nm resonance radiation is σ^+ circularly polarized and is incident along the direction of the external magnetic field, the depolarization effect vanishes, and in fact the higher cross section for photoionization from the $2P$ state can be used to produce a more intense source of polarized electrons. Moreover, if σ^- radiation is allowed to irradiate the ensemble of atoms for ~20 µsec prior to ionization, the $2S$ $m_J = -\frac{1}{2}$ state is optically pumped, thereby reversing the electron polarization. We calculate that with a cw dye laser generating ~1 W of power at 670.8 nm with a 0.1-Å bandwidth,[14] a factor of 10 increase in the polarized electron beam intensity to ~10^{10} e/pulse can be obtained using the existing atomic beam and flash-lamp configuration. An alternative technique of excitation of the $2P$ state might use a single-moded laser with its beam transverse to the Li beam to selectively populate the $2P_{3/2}$ $m_J = +\frac{3}{2}$ level.[15] The electronic polarization of the Li atoms might be reversed efficiently by the use of radio-frequency transitions without changing the static magnetic field in the photoionization region.[16]

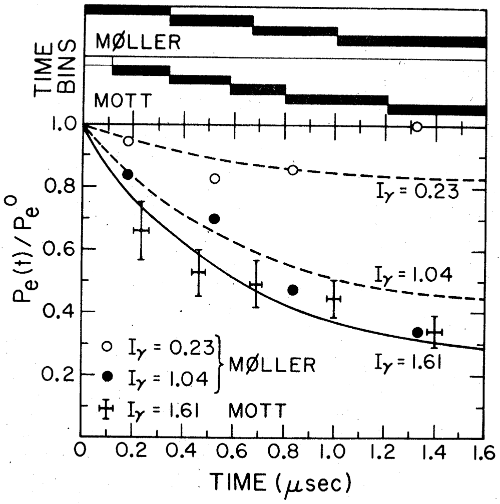

FIG. 2. Time dependence of electron polarization during flash lamp pulse. All Møller measurements have polarization uncertainties of ±0.24. Data points are plotted at the center of the time bins shown at the top of the figure. The horizontal error bars reflect the uncertainty in the determination of time $t = 0$. For the Møller measurements, this uncertainty is negligible. No Mott data point was obtained for $t < 114$ nsec because of excessive electronic noise.

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