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## POLARIZATION CORRELATION OF PHOTONS EMITTED IN AN ATOMIC CASCADE\*

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We describe observations of the correlation in linear polarization of two successive photons emitted in the cascade  $6^1S_0 \rightarrow 4^1P_1 \rightarrow 4^1S_0$  in calcium.

For the linear polarization correlation of two photons emitted simultaneously in the annihilation of singlet positronium, the odd intrinsic parity of the electron-positron pair implies a coincidence rate proportional to  $[1 - (\hat{\epsilon}_1 \cdot \hat{\epsilon}_2)^2]$ , where  $\hat{\epsilon}_1$  and  $\hat{\epsilon}_2$  are unit vectors in the directions of the photon linear polarizations.<sup>1</sup> This dependence was verified by observation of the azimuthal asymmetry in Compton scattering of the 0.5-MeV  $\gamma$  rays.<sup>2,3</sup> In the present case the photons  $\gamma_1$  and  $\gamma_2$  are emitted successively, and their wavelengths are in the visible range ( $\lambda_1 = 5513 \text{ \AA}$ ,  $\lambda_2 = 4227 \text{ \AA}$ ). It is therefore possible to detect the polarization correlation by means of conventional photomultipliers and linear polarizers of the Polaroid type. Since the initial and final atomic states have zero total angular momentum and the same parity, the correlation is expected to be of the form  $(\hat{\epsilon}_1 \cdot \hat{\epsilon}_2)^2$ . This experiment, like others involving correlations of two or more radiations, is of interest as an example of a well-known problem in the quantum theory of measurement, first described by Einstein, Podolsky, and Rosen<sup>4</sup> and elucidated by Bohr.<sup>5</sup>

The relevant levels of calcium and a sketch of the experimental apparatus are shown in Fig. 1. A tantalum oven produces an atomic beam of calcium. Its operating temperature

is about  $1000^\circ\text{K}$ , at which the beam density in the interaction region is  $3 \times 10^{10}$  atoms/cm<sup>3</sup>. Excitation radiation is generated by a low-voltage H<sub>2</sub> arc lamp, which emits an intense ultraviolet continuum. The interference filter for the lamp transmits a 300- $\text{\AA}$  band centered at  $2275 \text{ \AA}$ , corresponding to the transition  $4^1S_0 \rightarrow 6^1P_1$ . We use the H<sub>2</sub> lamp instead of a Ca resonance lamp because the latter would produce a very large undesirable 4227- $\text{\AA}$  background which could not be removed effectively by available filters. Since Doppler broadening reduces the absorption coefficient at the center of a resonance line, we orient the calcium beam

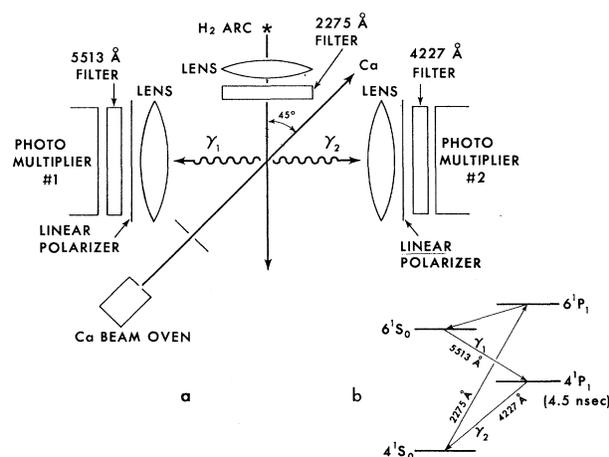


FIG. 1. (a) Schematic diagram of apparatus. (b) Level scheme for calcium.

at an oblique angle to the observation axis. This arrangement allows moderately high beam densities without appreciable trapping and multiple scattering of  $\gamma_2$  photons. The photomultiplier tubes were chosen for high quantum efficiency, low dark noise, and good time resolution. Photomultiplier No. 1 (RCA type 7265) has an S-20 cathode, with quantum efficiency  $Q \approx 10\%$  at 5513 Å; No. 2 (RCA type 8575) has a bi-alkali cathode, with  $Q \approx 20\%$  at 4227 Å. Narrow-band interference filters, centered at these wavelengths, are mounted in the detector assemblies. Each filter has a peak transmission  $T \approx 30\%$ . The over-all efficiency of each detector is  $\eta = QT\Omega/4\pi \approx 10^{-3}$ , where  $\Omega$  is the solid angle subtended by the detector.

The calcium atoms are excited from the  $4^1S_0$  ground state to the  $6^1P_1$  state ( $\lambda = 2275$  Å) and also, to some extent, to higher  $^1P_1$  states. From a Bates-Damgaard calculation of the transition rates<sup>6</sup> we estimate that about 10% of the atoms which do not return directly to the ground state go to the  $6^1S_0$  level, which is the initial state for the cascade. Since most of the remaining excited atoms reach the  $4^1P_1$  level by other routes, the rate  $R$  of  $\gamma_2$  emission ( $R \approx 10^6$ /sec) is about ten times the rate for the desired cascade. The coincidence rate (with polarizers removed) is  $0.1\eta_1\eta_2R \approx 10^{-1}$ /sec. To reduce photomultiplier dark noise we cool the tubes to  $-5^\circ\text{C}$ . In addition, carefully placed baffles are necessary to reduce the pickup of light from the oven and of wall fluorescence due to ultraviolet  $\text{H}_2$  radiation.

Time analysis permits a direct display of the coincidence rate as a function of delay time. Pulses from photomultipliers No. 1 and No. 2 are fed into the start and stop inputs, respectively, of a time-to-height converter. Its output pulse amplitude, proportional to the delay time between the  $\gamma_1$  and  $\gamma_2$  pulses, is analyzed by a multichannel pulse-height analyzer. An asymmetry is expected in the time correlation because the  $4^1P_1$  intermediate state decays exponentially. (Its mean life is about  $4.5 \times 10^{-9}$  sec.<sup>7</sup>) The instrumental time response, measured by means of a radioactive source and fast scintillator, has a full width at half-maximum of about  $6 \times 10^{-9}$  sec. In the case of a cascade having a longer lived intermediate state, the lifetime can be measured directly from the time-correlation curve. (See Kaul<sup>8</sup> for a description of such observations on an excited state of Hg.)

The singles rates from individual photomultipliers vary insignificantly with polarizer orientation; the small variation observed is due to polarization of stray light reflected within the vacuum chamber. Measurements with the polarizer axes set parallel and perpendicular are made alternately; one polarizer is fixed, and the orientation of the other is changed by  $90^\circ$  every 15 minutes. We have made runs with different orientations of the fixed polarizer, obtaining in each case a correlation which depends only on the relative angle between the axes. The results of a 21-h run, shown in Fig. 2, indicate clearly the difference between the coincidence rates for parallel and perpendicular orientations. They are consistent with a correlation of the form  $(\hat{e}_1 \cdot \hat{e}_2)^2$ .

If a magnetic field is applied at the interaction region, an atom will precess during the time it spends in the intermediate state. If the field is perpendicular to the detector axis, an oscillatory function  $(1 + \cos^2\omega t)$ , where  $\omega$  is the Larmor frequency, will modulate the exponential decay function. This effect has been studied in nuclear angular-correlation experiments,<sup>9</sup> and should be observable here when the polarizers are removed.

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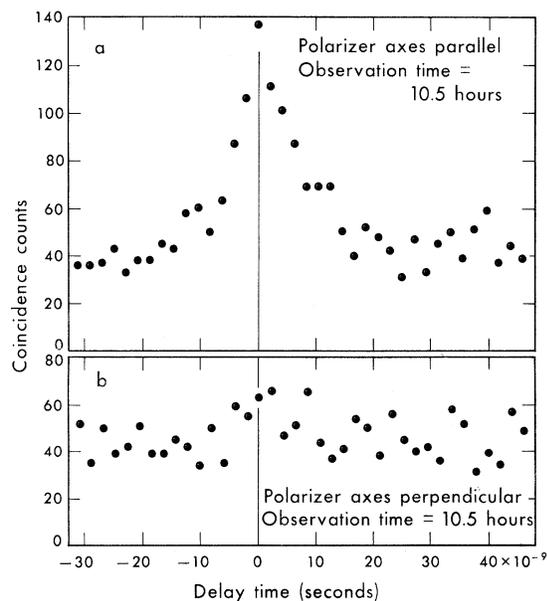


FIG. 2. Coincidence counts, as a function of relative time delay, showing polarization correlation. Each point represents a sum over three analyzer channels. Very slight peak in (b) is probably due to 6% transmission of crossed polarizers.

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## OFF-RESONANT LIGHT AS A PROBE OF OPTICALLY PUMPED ALKALI VAPORS\*

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Vapors of optically pumped atoms are usually studied by observing the interaction of resonant light with the atoms. Experiments using off-resonant light have been proposed and have been carried out with different isotopes of mercury.<sup>1</sup> In this Letter we discuss the fundamental criteria which determine the feasibility of experiments using off-resonant light, and we show explicitly which observables can be measured in the important case of the alkali atoms. The light of a Sr II line is shown to be a good nonperturbing probe for optically pumped rubidium atoms.<sup>2</sup>

Consider a vapor of alkali atoms with nuclear spin  $I$ , traversed by a beam of monochromatic light whose frequency  $\omega$  almost coincides with a resonant absorption frequency  $\omega_F$  of the vapor. We will assume that the hyperfine structure (hfs) in the optical line of the alkali atom is chiefly due to the splitting of the ground-state  $^2S_{1/2}$  level into the two hfs sublevels  $F = I \pm \frac{1}{2}$ . Then one can show that the indices of refraction for right (+) and left (-) circularly polarized light, propagating in the direction of the unit vector  $\hat{i}$ , are

$$n_{\pm} = 1 + \delta n_0 \pm \delta n_1, \quad (1)$$

where

$$\delta n_0 = \sum_F \frac{Ne^2\lambda_F f_J(\omega - \omega_F)}{2mc[(\omega - \omega_F)^2 + \Gamma^2]} p_F, \quad (2)$$

and

$$\delta n_1 = \sum_{F^-} \frac{Ne^2\lambda_F f_J(\omega - \omega_F)}{2mc[(\omega - \omega_F)^2 + \Gamma^2]} \times \left[ \frac{11 - 4J(J+1)}{4} \right] \frac{\langle \vec{\mu} \rangle_{F^-} \cdot \hat{i}}{g_J \mu_0}. \quad (3)$$

Here  $N$  is the atomic density,  $\Gamma$  is the collision-broadened width of the excited state,  $f_J$  is the absorption oscillator strength for the  $^2P_J$  resonance line, and  $\lambda_F$  is the wavelength of the transition. The index of refraction  $n_{\pm}$  depends on the ground-state observables  $p_F$  and  $\langle \vec{\mu} \rangle_{F^-}$ , where  $p_F$  is the probability that the atom is in the hyperfine sublevel  $F$ , and  $\langle \vec{\mu} \rangle_{F^-}$  is the expectation value of the electron magnetic moment within that sublevel.<sup>3</sup> Thus all of these observables may be detected using off-resonant light.

An examination of Eqs. (2) and (3) shows that the wavelength  $\lambda$  of the off-resonant probing light must fulfill the following criteria. In order to detect the relative hfs populations  $p_F$ , it is necessary that the wavelength of the probing light be much closer to one hfs component,  $\lambda(F)$ , of an absorption line than to the other,  $\lambda(F')$ ; i.e.,  $|\lambda - \lambda(F)| \ll |\lambda - \lambda(F')|$ . If this is not the case,  $\delta n_0$  [Eq. (2)] may be expanded in a power series  $\delta n_0 \propto 1 + \beta_1(p_F - p_{F'})[\lambda(F) - \lambda(F')]/\{\lambda - \frac{1}{2}[\lambda(F) + \lambda(F')]\} + \dots$  in which the terms containing  $p_F - p_{F'}$  are negligibly small. However, the net magnetization  $\langle \vec{\mu} \rangle_F + \langle \vec{\mu} \rangle_{F'}$  can still