sion lifetimes of nuclides in the mass-300 region.

There remain, however, large uncertainties in the fission lifetimes of superheavy elements because of uncertainties in the neutron single-particle spectrum at $N = 184$, particularly in the energies of the highly degenerate orbitals $h_{11/2}$, $i_{13/2}$, and $k_{17/2}$. The observation of the neutron single-particle states $\frac{1}{2}^-[761]$, $\frac{1}{2}^-[750]$, and $\frac{1}{2}^+[880]$ in the mass-250 region would resolve this problem.

In conclusion, we have observed the proton orbital $\frac{1}{2}^-[521]$ in the mass-250 region. The position of this level gives a value of ~1.5 MeV for the $f_{7/2}$-$f_{5/2}$ splitting and a proton shell correction of ~ −2.8 MeV at $Z = 114$.

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Observation of Two-Photon Optical Free-Induction Decay in Atomic Sodium Vapor

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We have used the Stark-switching technique to observe two-photon optical free-induction decay in the visible using sodium vapor. Transients were observed with use of both a resonant and a nonresonant intermediate state for the two-photon transition.

Since the first observations[1] of photon echoes many single-photon coherent optical transients have been measured.[2] The extension of the techniques of coherent optical transients to two-photon transitions has recently become of interest. These techniques lead to spectroscopic studies in the time domain which are complementary to those made in the frequency domain. We report in this Letter the first use of the Stark-switching technique[3] to observe two-photon optical free-induction decay (FID). These measurements were made in the visible with use of sodium vapor, and the transients occurred on a nanosecond time scale. Two situations were studied: Simple two-photon FID was observed when the intermediate state for the two-photon transition was nonresonant. However, when the intermediate state was resonant, a different behavior was seen. This latter situation has not previously been studied experimentally and our results presently are not fully understood.

The interest in two-photon coherent effects began when Hartmann first discussed the possibility of observing Raman echoes.[4] These Raman echoes have only recently been observed by Hu, Geschwind, and Jedju.[5] Shoemaker and Brewer have observed a coherent transient phenomenon known as Raman beats.[6] Detailed theoretical analysis of Raman beats has been made[7-9] and the close relationship to two-photon free-induction decay has been discussed.[7] Other experimental work on two-photon transients include the reported observation of two-photon self-induced transparency[10] and measurements of the decay of two-

photon coherence[11] by examining the generation of ultraviolet light by a delayed nonresonant picosecond pulse following a two-photon resonant pulse. The most recent experimental work is the observation of two-photon optical mutation and free-induction decay in the infrared region of the spectrum.[12] The theory of two-photon coherent transients has attracted great attention and many theoretical aspects have been discussed.[7-9,13,14]

In our experiments, two single-axial-mode cw dye lasers were operated at different wavelengths ($\lambda_2 \sim 590$ nm and $\lambda_1 \sim 569$ nm) and the beams from them were passed in opposite directions through a 2.5-cm-long Pyrex cell containing atomic sodium vapor at a density of about 10^{12} cm^{-3}. The beams were focused between two Stark electrodes (~ 2-mm spacing) in the cell; the 590-nm (569-nm) beam was focused with a 50-cm- (100-cm-) focal-length lens to a spot with radius of about 10^{-2} cm (2×10^{-2} cm). A dc Stark field of 2.5 kV/cm was applied so that the $4D_{3/2}$ ($m_J = \pm\frac{3}{2}$) states could be selectively excited. The laser frequencies ω_1 and ω_2 were adjusted such that ω_2 ($\lambda_2 \sim 590$ nm) was resonant or nearly resonant with the $3S_{1/2}$–ground-state to $3P_{1/2}$–intermediate-state transition; and for all experiments reported here, the sum frequency $\omega_1+\omega_2$ was approximately centered on the $3S_{1/2}$ ($F=1$) $\rightarrow 4D_{3/2}$ ($m_J = \pm\frac{3}{2}$) two-photon transition. After passing through the cell and a chopper, the 590-nm laser beam was focused on a fast silicon photodiode (Lasermetrics model 31-17), the output of which was amplified (A.R. model 1W-1000) and displayed on a Tektronix 7904 oscilloscope. (Overall bandwidth of the detection system was estimated to cover the frequency range 1–300 MHz.) The chopper was required to minimize the average current drawn by the photodiode.

The FID signals were generated by applying an additional voltage in the form of a step of approximately 160 V across the Stark plates. This step voltage caused the $4D_{3/2}$ ($m_J = \pm\frac{3}{2}$) levels to suddenly shift (within 2 nsec) by approximately 120 MHz. The shift in energies of the $3P_{1/2}$ and $3S_{1/2}$ ($F=1$) states which is directly a result of the Stark field is negligible.

Figure 1 illustrates the Stark pulse and the two-photon FID signal obtained with ω_2 tuned 6 GHz off the $3P_{1/2}$–intermediate-state resonance. This mistuning is approximately 6.5 Doppler half-widths off the intermediate-state resonance. The signal appears as a beat note, as predicted in Ref. 7, which is the result of interference between the laser radiation at ω_2 and the radiation

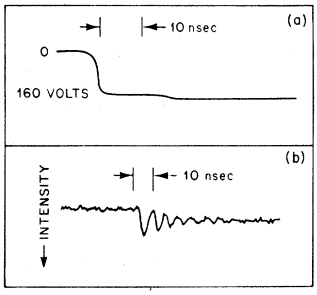

FIG. 1. (a) Oscilloscope trace of the voltage pulse applied to the Stark plates. (b) Two-photon free-induction decay observed with a nonresonant intermediate state where the emission and laser produce a 120-MHz beat. (Note the difference in the time scales.)

from the two-photon coherently prepared atoms. This signal is in many respects similar to the well-known FID signals obtained by Stark switching in single-photon transitions.[15] An optical-nutation signal was not observed because the use of oppositely propagating beams produced a Doppler-width reduction such that the Stark pulse shifted the $4D_{3/2}$ ($m_J = \pm\frac{3}{2}$) levels completely out of resonance. The two-photon FID can be visualized, with use of the vector model,[7,14] as the precession in a rotating frame of the two-photon "polarization" vector $\vec{r}$ about the effective field $\vec{\gamma}$.[16] The components of $\vec{\gamma}$ are $\gamma_1 = -\kappa\mathcal{E}_1\mathcal{E}_2$, $\gamma_2 = 0$, and $\gamma_3 = \delta + (\Delta E_1 - \Delta E_2)/\hbar$, where $\kappa = |P_{1n}P_{n2}/2\hbar^2\Delta|$, $\mathcal{E}_1$ and $\mathcal{E}_2$ are the optical electric-field amplitudes, P_{ij} are transition dipole moments, $\Delta E_i = -|P_{in}\mathcal{E}_i|^2/4\hbar\Delta$ are the optical Stark shifts,[17] $\Delta \simeq \Omega_{n2} - \omega_2$, and Ω_{n2} is the frequency separation between the intermediate and ground states. The mistuning from two-photon resonance is given by

$$|\gamma_3| = \Omega_{12} - (\omega_1+\omega_2) + (\omega_1-\omega_2)v/c + (\Delta E_1 - \Delta E_2)/\hbar,$$

where Ω_{12} is the frequency separation between the excited state (1) and ground state (2), and v is the atomic velocity along the direction of the beams. These equations are identical to those for the single-photon vector model with the single-photon Rabi frequency replaced by $\kappa\mathcal{E}_1\mathcal{E}_2$ and the addition of the optical Stark shifts. Hence we can make use of results already obtained for single-photon FID[9,15] which give a solution in terms of a Doppler-averaged integral. We have evaluated this integral in closed form, and we find the time-dependent part of intensity at the detector

to be

$$I(t) \propto -\mathrm{Re}(\kappa\mathcal{E}_1\mathcal{E}_2)^2 \exp[-(i\Delta_s + 1/T_2)t]\{(1+\eta)\exp[(\alpha+i\gamma_3/\sigma)^2+2\alpha\tau]\,\mathrm{erfc}(\alpha+\tau+i\gamma_3/\sigma) - (1-\eta)\exp[(\alpha-i\gamma_3/\sigma)^2-2\alpha\tau]\,\mathrm{erfc}(\alpha-\tau-i\gamma_3/\sigma)\}, \quad (1)$$

where $\eta = (1+\kappa^2\mathcal{E}_1{}^2\mathcal{E}_2{}^2T_1T_2)^{-1/2}$, $\alpha = 1/\eta\sigma T_2$, $\tau = \frac{1}{2}\sigma t$, $\sigma^2 = 2kT(\omega_1-\omega_2)^2/mc^2$, and Δ_s is the shift of the two-photon transition frequency produced by the Stark pulse. In agreement with earlier work,[9,15] this equation predicts an initial transient (which might not be observed in our experiment[18] because of the finite risetime of the Stark pulse), followed by a damped sine wave. The decay time of the damped sine wave is $\eta T_2/(1+\eta)$. For the conditions in Fig. 1(b), $\kappa\mathcal{E}_1\mathcal{E}_2 \simeq 2\times10^7$ sec^{-1} and $T_2 = 51$ nsec and $T_1 = 34$ nsec (these times are shorter than those determined by the natural lifetime, 52 nsec, of the $4D$ states because of the transit time of an atom across the beam); we obtain a damping time of 15 nsec, in reasonable agreement with Fig. 1(b). Spatial inhomogeneities in the Stark pulse field may also contribute to an additional decrease in the damping time.

In the case of a resonant intermediate state ($\omega_2 \simeq \Omega_{n2}$), the vector model used above is no longer valid and the phenomena can be complicated by single-photon transients. However, at sufficiently low intensity in the two beams ($I_1 \lesssim 5$ W/cm^2, $I_2 \lesssim 2$ W/cm^2), we have found FID signals which are essentially the same as Fig. 1(b). At higher intensities, the coherent transient signals are quite different. Figure 2 shows two typical transients. The intensity of the ω_1 beam was ~ 32 W/cm^2 and is the same for both (a) and (b). The intensity at ω_2 was ~ 4 W/cm^2 in 2(a) and ~ 17 W/cm^2 in 2(b). At the intensities for which Fig. 2(a) was taken, an excitation spectrum, which was obtained by monitoring the fluorescence from the $4D$ states while scanning ω_1 (with ω_2 fixed), displays a well-resolved ac Stark splitting[19] of ~ 80 MHz. The magnitude of the splitting was found to be proportional to $I_1{}^{1/2}$ and approximately

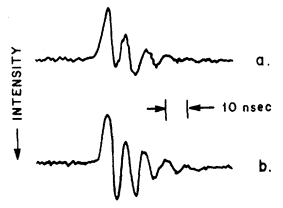

FIG. 2. Two-photon free-induction decay observed with a resonant intermediate state. For both parts of the figure, $I_1 \approx 32$ W/cm^2; for (a) $I_2 \approx 4$ W/cm^2 and for (b) $I_2 \approx 17$ W/cm^2.

equal to $P_{1n}\mathcal{E}_1/h$. In the case of Fig. 2(b), I_2 was sufficient to power broaden the spectra so that the ac Stark splitting was unresolved, i.e., $P_{n2}\mathcal{E}_2/2h \geq P_{1n}\mathcal{E}_1/h$. In each case the FID transient appears as a beat note whose frequency is determined only by the amplitude of the Stark pulse. The beat frequency is essentially independent of the intensitites of either laser beam.

While the transient in Fig. 2(b) is basically only a damped sine wave, Fig. 2(a) contains at least one other component. This other component is tentatively ascribed to a single-photon optical-nutation[3,20] signal which arises when the $3P_{1/2}$ level is slightly shifted and a new velocity group suddenly becomes resonant with ω_2. This shift of the $3P_{1/2}$ state is indirectly caused by the Stark pulse in the following way. The Stark pulse directly causes a shift of the $4D_{3/2}$ level; and this, in turn, results in a change in the optical Stark shift of the $3P_{1/2}$ level caused by light at ω_1. If the new velocity group which is shifted into resonance was not previously saturated, then an optical-nutation signal will appear. For sufficiently large values of I_2 [as in Fig. 2(b)], power broadening causes the new velocity group to be already saturated so that no optical-nutation signal is obtained. The disappearance of the ac Stark splitting in the excitation spectrum is consistent with this saturation. Larger values of I_1 produce increased splittings and required correspondingly larger values of I_2 to obtain the simple damped-sine-wave behavior of Fig. 2(b). A series of experiments was also performed with I_1 at 32 W/cm^2 for various values of I_2. Once I_2 was sufficiently high that the signal appeared as a simple damped sine wave, further increases in I_2 only caused the signal to decay more rapidly as a wider two-photon line is excited. The phase of the oscillations in Fig. 2 is clearly different from that in Fig. 1(b). This change in phase is not fully understood.

We have reported here observations of two-photon optical transients in the visible region of the spectrum. These measurements have been made with the Stark-switching technique on a nanosecond time scale. As in the single-photon case, this technique has the advantages of high sensivitivy due to heterodyne detection and the

ability to tailor the excitation pulses electronically. Samples which do not exhibit large Stark effects can be studied with laser-frequency switching.[21] By using a resonant or nearly resonant intermediate state to enhance greatly the two-photon transition,[22] large signals are easily obtained. However, more theoretical study is required for a complete understanding of the phenomena observed in the case of a resonant intermediate state.

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Effective-Photon Theory: A Critical Analysis[(a)]

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It is shown that effective-photon theory is in serious conflict with established experimental results in two areas. A lower limit on the total photon-photon scattering cross section is derived which requires this cross section to be unacceptably large. The rate of beam degradation also is examined and shown to be too high by many orders of magnitude.

In a recent series of articles,[1-3] Panarella has introduced the concept of the effective photon in order to explain certain results of laser-induced gas breakdown. It is postulated that energy may be exchanged between photons according to the following inelastic process[4]:

$$\gamma(\vec{p}) + \gamma(\vec{q}) \rightarrow \gamma(\vec{p}') + \gamma(\vec{q}'). \quad (1)$$

The essential feature of effective-photon theory is that because of this inelastic photon-photon scattering, an intense beam of monochromatic radiation is rapidly degraded to a beam of large frequency width, creating a large number of high-energy photons. These high-energy photons then ionize the gas by a single-photon process; they