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Observation of optical nutation in a collinear fast-ion-beam–laser experiment

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We report the first observation of optical nutation in a collinear fast-ion-beam–laser experiment. 35-keV Ba^+ ions are suddenly tuned into resonance with a strong laser field by post acceleration over a short distance. The degree of excitation as a function of time is monitored by observing the spontaneously emitted light along the ion beam. Oscillations as a function of transition, laser intensity, and laser frequency detuning have been studied. Theoretical curves have been calculated in the density-matrix formalism.

Several experimental techniques have been developed during the last two decades in order to study optical coherent transients. These have been generated by bringing the sample in or out of resonance with the laser by switching either the laser field amplitude,[1–3] the atomic levels—by Stark pulses,[4,5] the laser frequency[6–8] or the relative phase of the laser and the sample polarization.[9] In this Rapid Communication we report the first observation of optical nutation in a fast-ion-beam–laser experiment with a Doppler-shift technique that should be useful in the study of coherent optical transients in atomic ions.

By introducing a step in the electric potential along the common beam path in a collinear fast-ion-beam–laser experiment, the frequency of the laser light can be Doppler shifted with respect to an atomic transition frequency in a time interval shorter than the decay time of the atomic phase coherence. Due to the inherent time resolution in fast-beam experiments, this offers the possibility to observe optical transient effects and has been used to study various transient interference phenomena like the optical analogue of Ramsey interference fringes.[10,11] The feasibility to observe optical nutation in such experiments was recognized already in 1977,[12] but has, to our knowledge, not been demonstrated up to now.

Figure 1 shows part of the experimental arrangement: An isotopically pure beam of ions is overlapped with the counterpropagating radiation from a single-mode cw ring dye laser (CR-699) with an effective bandwidth less than 1 MHz. The ions are exposed to a "square pulse" of resonant radiation by tuning the Doppler-shifted frequency experienced by the ions in a Faraday cage at nonzero potential to an ionic resonance frequency. In this work, transitions are induced between levels in the $5d$ and $6p$ configurations (see Fig. 2) in singly ionized barium (^{138}Ba with nuclear spin $I=0$). The degree of excitation of the upper level taking part in a transition is monitored by measuring the intensity of the spontaneously emitted, incoherent, light from this level. This restricts the class of observable optical transient effects, but in this way one may observe time-resolved population variations simply by moving the light collector along the ion beam as well as frequency-resolved variations at any given time after the pulse onset by scanning the laser frequency with the light collector at a fixed position. The square-pulse excitation is also convenient for the study of the influence of laser-field amplitude and frequency detuning on the period of optical nutations.[3]

However, some crucial prerequisites must be fulfilled:

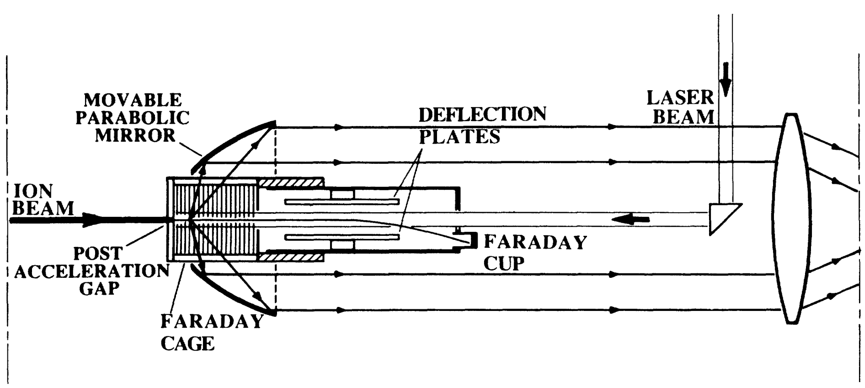

FIG. 1. Part of the experimental setup. Inside the Faraday cage the ions are Doppler tuned into resonance with the counterpropagating laser radiation. A time-resolved signal is obtained by moving the reflector along the ion beam.

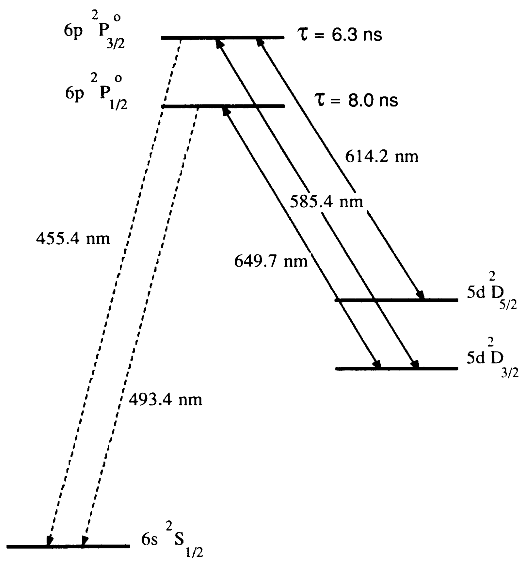

FIG. 2. Schematic level scheme of Ba II indicating lifetimes and the transitions used.

(i) The square-pulse rise time, i.e., the ion post acceleration time, must be short compared with the lifetime of the upper level and the period of nutation. (ii) The time resolution, i.e., the spatial resolution of the light-collecting system, must be so high that the nutation pattern will not be smeared out. Also, the light-collection system must have a high efficiency since low ion currents through small apertures have to be used. (iii) To prevent a broad distribution of nutation frequencies, the electric field amplitude of the laser light over the interacting volume of the ion beam should be as constant as possible and the longitudinal velocity distribution of the ions should be sufficiently narrow. (iv) The Rabi frequency, which increases with increasing laser-field amplitude and transition probability, must be of the order of or greater than the linewidth of the transition.

The square pulse risetime is determined by the ion velocity and the smoothly changing electrostatic potential of 500 V between the entrance plate of the Faraday cage and the earthed plate in front of it. In this experiment, the time to switch the whole velocity distribution into resonance is about 1.5 nsec.

The time resolution is determined by the optics of the photon counting system[13] which has been designed to give high efficiency at the same time as high resolution. The system consists of (see Fig. 1) a movable parabolic reflector, a converging lens, a broad-band monochromator, and a photomultiplier tube. The reflector collects fluorescence light from its focus with an acceptance angle of about 10% of the total solid angle and directs it to the photomultiplier tube through the lens and the monochromator. For a 0.5-mm-diam beam of 35-keV Ba^+ ions, the time resolution is 1.3 nsec.[13]

The ion beam is collimated to a diameter of 0.5 mm and the spotsize of the Gaussian laser beam inside the Faraday cage is 2.4 mm. This reduces the variation of the laser-field amplitude over the ion beam (cf. Ref. 14), on which the laser beam is centered, to less than 1%. The compression of the longitudinal velocity distribution in a fast-ion beam[15,16] is essential for the experiment. The Doppler contribution to the observed linewidth at low laser power (less than 1 mW) has been determined to 35 MHz, which is sufficiently narrow to allow the observation of several periods of oscillations before this inhomogeneous dephasing mechanism has smeared out the nutation pattern.

Finally, the laser power densities required for nutation periods in the nsec range are easily obtained with the ring dye laser for the transitions studied here, producing Rabi frequencies of the order of 50–100 MHz, to be compared with the homogeneous linewidths of 20–25 MHz.

Our description of the observed nutations is based on the density-matrix formalism. Considering an idealized two-level atom where spontaneous emission has been neglected, this model yields[17] that the populations will oscillate with an angular frequency Ω which is given by

$$\Omega^2 = (\mathbf{D}_{1,2}\cdot \mathbf{E}_0)^2 + (\omega-\omega_0)^2 .$$

Here $\mathbf{D}_{1,2}$ is the matrix element of the dipole operator, divided by Planck's bar constant, and $\mathbf{E}_0$ the amplitude of the laser field. The second term is the squared detuning between the frequency of the laser field ω and the resonance frequency ω_0, and in the case of zero detuning Ω equals the Rabi frequency $\mathbf{D}_{1,2}\cdot \mathbf{E}_0$.

In our experiment each of the two levels is degenerate, corresponding to different orientations of the dipole moment, denoted by the magnetic quantum number m. Furthermore, the spontaneous emission is substantial. These effects have to be included in any attempt to reproduce experimental data. It should be noted that in a fast-ion-beam experiment, because of the low density of the ion beam, the only important homogeneous dephasing mechanism is due to the radiative damping.

The laser induces transitions between two degenerate levels which we denote 1 and 2, 1 being the lower level, which is metastable. Since the laser light is linearly polarized, there will be only $\Delta m = 0$ transitions. The population density of the $2m$ states, $\rho_{2m,2m}$, will oscillate with different Rabi frequencies since the dipole moment $\mathbf{D}_{1m,2m}$ depends on the magnetic quantum number m.[18] Furthermore, the spontaneous decay causes not only damping of the oscillations, but also a coupling between the quantum numbers m. In the rotating wave approximation, the equations of motion of the density-matrix elements take the form

$$\frac{d\rho_{2m,2m}}{dt} = -\tfrac{1}{2}iV_m^*\tilde{\rho}_{1m,2m} + \tfrac{1}{2}iV_m\tilde{\rho}_{2m,1m} - 2\gamma\rho_{2m,2m} ,$$

$$\frac{d\rho_{1m,1m}}{dt} = \tfrac{1}{2}iV_m^*\tilde{\rho}_{1m,2m} - \tfrac{1}{2}iV_m\tilde{\rho}_{2m,1m} + 2\sum_{m'}\gamma_{m'm}\rho_{2m',2m'} ,$$

$$\frac{d\tilde{\rho}_{1m,2m}}{dt} = \tfrac{1}{2}iV_m(\rho_{1m,1m} - \rho_{2m,2m}) - (i\Delta+\gamma)\tilde{\rho}_{1m,2m} ,$$

$$\frac{d\tilde{\rho}_{2m,1m}}{dt} = -\tfrac{1}{2}iV_m^*(\rho_{1m,1m} - \rho_{2m,2m}) + (i\Delta-\gamma)\tilde{\rho}_{2m,1m} ,$$

where Δ is the detuning, γ the total decay rate from either of the states $2m$, $\gamma_{m'm}$ the decay rate of the particular

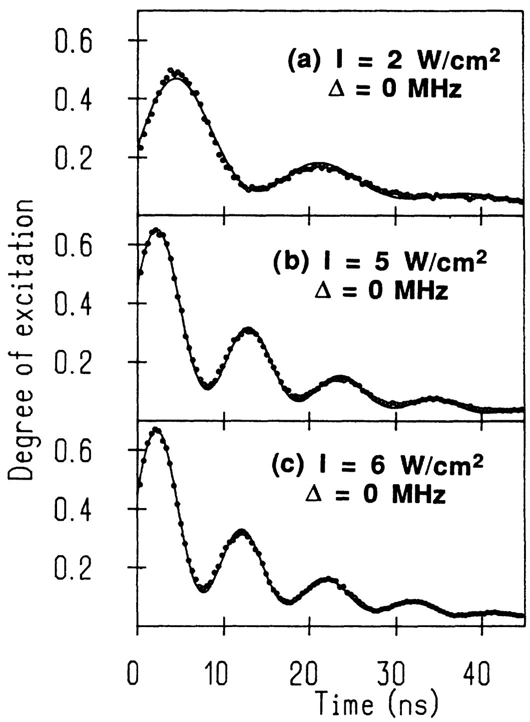

FIG. 3. Degree of excitation as a function of time for three different intensities. The $5d\,^2D_{3/2}$–$6p\,^2P^o_{1/2}$ transition has been used. The solid line is the calculated curve.

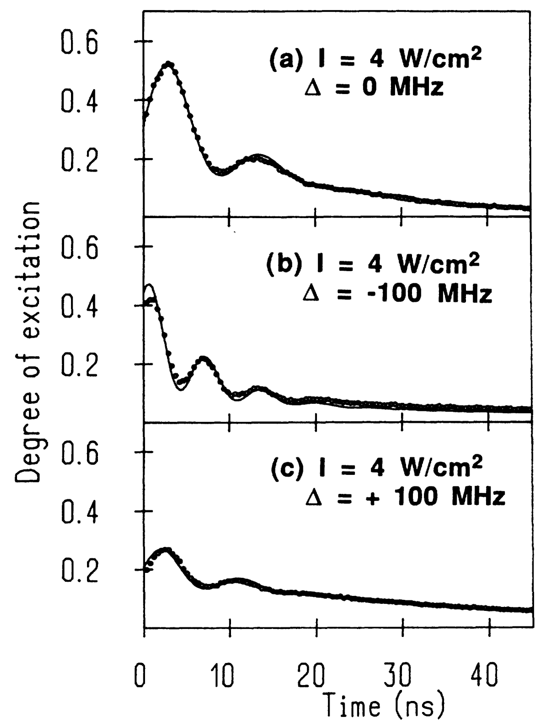

FIG. 4. Degree of excitation as a function of time for three different detunings. The $5d\,^2D_{5/2}$–$6p\,^2P^o_{3/2}$ transition has been used. The solid line is the calculated curve.

transition $2m'$-$1m$, and

$$\mathbf{V}_m = \mathbf{E}_0 \cdot \mathbf{D}_{1m,2m} ,$$

$$\tilde{\rho}_{1m,2m} = e^{-i\omega t} \rho_{1m,2m} ,$$

$$\tilde{\rho}_{2m,1m} = \tilde{\rho}^*_{1m,2m} = e^{i\omega t} \rho_{2m,1m} .$$

Experimental curves have been produced with many parameters being varied, such as the polarization of the laser light, the detuning as seen by the ions in the beam, the laser field amplitude, and the transition used. Figures 3 and 4 show observed optical nutations under varying experimental conditions as well as least-squares-fitted nutation curves for a few cases. The effect of the velocity distribution of the ions and the time resolution have been taken into account as well as the post acceleration potential function which is needed to assign initial values of the density matrix. The values of the dipole moments have been taken from the literature[19] and have been kept fixed in the calculations, whereas the laser-power density and the detuning have been allowed to vary to obtain a good fit. This procedure is motivated by the relatively large uncertainties in laser-power density ($\approx$ 20%) and detuning ($\approx$ 20 MHz, caused mainly by drifts in the acceleration voltage) compared with the uncertainty of the dipole moments (typically 10%). Within these error limits, the calculations reproduce the observations very well.

The two zero detuning curves of Fig. 4(a) and, e.g., Fig. 3(b) illustrate the difference between cases with only one Rabi frequency [Fig. 3(b)] and with two Rabi frequencies present [Fig. 4(a)], which interfere and smear out the oscillation pattern.

An interesting feature is the asymmetry in behavior between a positive and a negative detuning, which is seen in Figs. 4(b) and 4(c). The explanation of this phenomenon is that when the detuning is negative, many ions will be postaccelerated across the resonance before measurements start, but in the case of positive detuning, this will not happen. This creates different initial conditions for the two cases and hence an asymmetry.

We have demonstrated the possibilities that the rapid Doppler-shift technique in collinear fast-ion-beam-laser spectroscopy offers in studying optical transient effects. With commercially available power meters with absolute accuracies on the order of 1% this method may be possible to use to measure transition probabilities with an accuracy of a few percent.

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