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Suppression of Cascade Effects in Beam-Foil Mean-Life Measurements*

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Using the spatial decay of quantum beats, we have succeeded in suppressing optical cascading effects in the mean-life measurements of levels in foil-excited O II ions. This straightforward technique may be applied to any excited level with total angular momentum $J > \frac{1}{2}$ and a lifetime within the range of the beam-foil technique. With the present method, mean lives are obtained in terms of the Landé g factors and an external magnetic field, instead of the customary particle speed and decay length.

Mean lives and g factors of excited levels of ions and atoms can be extracted from polarization changes of the emitted light as a function of uniform external magnetic field strength. The beam-foil excitation process produces not only the requisite excited-level alignment, but also a sharp origin in space (or time relative to excitation) for observation of coherence-dependent phenomena such as the Hanle effect and quantum beats.[1,2] The usual Hanle signal depends on the product $g\tau$ of the Landé g factor and the mean life τ of the level of interest,[3] although it is possible in principle to obtain both g and τ in a single measurement by observing the Hanle signal for a finite time interval.[1,4] We present here a new method of obtaining τ which is particularly suited to the beam-foil technique.[5] The important feature of the present method is that optical cascades do not strongly influence our results, in contrast to possible cascade-induced complications of the Hanle effect[6] or of the usual beam-foil spectroscopic (BFS) mean-life measurement method.[5]

Our measurement method is illustrated in Fig. 1(a). Fast ions with velocity v are excited by passage through a thin carbon foil. Light emitted from the ions is collected from a narrow region about a fixed point x_0 in space; this light, after transmission through a linear polarizer, is spectroscopically analyzed and detected. The foil is advanced at a uniform rate until it passes x_0; it is then retracted at the same rate until it has returned to the initial position. The detected light intensity increases as the foil approaches x_0, drops to a background level when the foil passes x_0, and follows a mirror image curve when the foil is retracted. The usual BFS mean life is obtained from such a measurement.

Let a fixed, uniform, external magnetic field H_0 be applied perpendicular to the plane formed by the beam and direction of observation during one half of the cycle; characteristic beats in the emitted light occur if the level is coherently aligned, as Fig. 1(b) shows. Writing t as the time of observation relative to excitation, the intensity of observed light is described by[2]

$$I(t,H_0) = e^{-t/\tau}[A + B\cos 2\omega_L t] + C(t,H_0) + D; \quad (1)$$

$\omega_L = g\mu_B H_0/\hbar$ is the Larmor frequency, D is any constant background, and $C(t,H_0)$ describes light which emanates from the level of interest by virtue of cascade population from higher excited levels. The difference between $I(t)$ without and

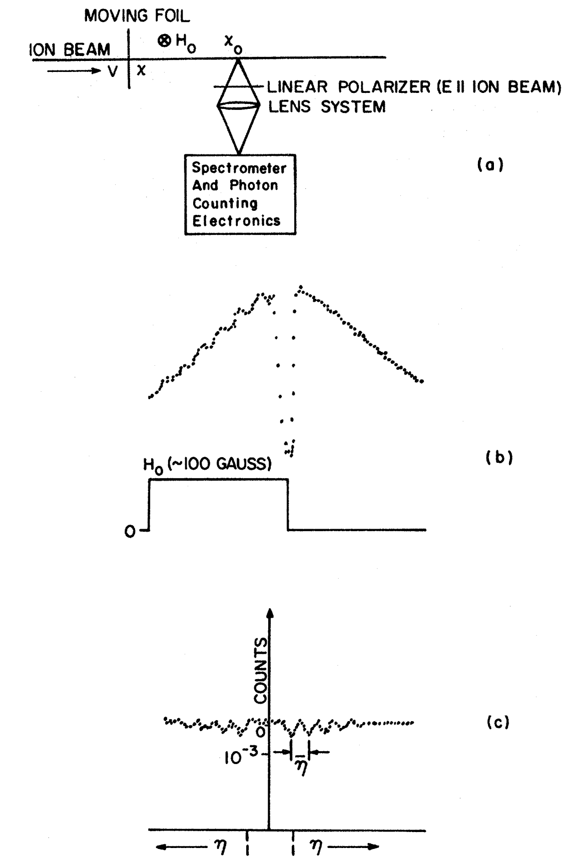

FIG. 1. (a) Schematic of experimental setup. The direction of the external magnetic field H_0 is into the paper. (b) Spatial decay of quantum-beat signal (left) and spatial decay curve with H_0 turned off (right). The transition was λ4590.97 from the $3p\,^2F_{7/2}^\circ$ level of O II. (c) The quantum-beat signals shown are accumulated over a time interval (about 30 sec per channel) of alternating modes of data collection. Each cycle with magnetic field H_0 on and the photons counted in an additive mode is followed by another cycle with H_0 off and photons counted in a subtractive mode. The same transition as in (b) was used. The distance traveled by the moving foil and the magnetic field strength H_0 are slightly different from those in (b).

with applied magnetic field is

$$\Delta I \equiv I(t,0) - I(t,H_0) = Be^{-t/\tau}[1-\cos 2\omega_L t] + C(t,0) - C(t,H_0). \qquad (2)$$

The contribution to ΔI arising from cascades comes only from the relative effects produced by the external field. An essential requirement for the appearance of the quantum beats is that the states involved be excited at the same instant of time; this occurs at the foil. However, cascade repopulation takes place without meeting that requirement. Therefore, those states which are populated by cascades do not contribute to the beat signal, except as incoherent background. One does expect a finite-time Hanle effect. However, the fixed external field we employ is large relative to that used for Hanle-effect work, so any Hanle-effect contribution to the observed light will be small. Furthermore, with the small light polarization usually observed in these measurements, such aligned cascade contribution has been found to be negligible; we have searched unsuccessfully for effects of strong cascades on quantum-beat (fixed t, variable H_0) periods.[7] Consequently, the approximation $C(t,0) - C(t,H_0) \equiv 0$ is not expected to decrease accuracy.

We set $2\omega_L t = a\eta$, where η is the channel number in a multiscaler display corresponding to the distance $x - x_0$. Then $I(t,H_0)$ can be rewritten as

$$I(\eta,H_0) = [A + B\cos a\eta] \times \exp(-a\eta/2\omega_L\tau) + C(\eta,H_0) + D, \qquad (3)$$

where $a = 2\pi/\bar{\eta}$ [see Fig. 1(c)] in terms of $\bar{\eta}$, the spatial period of the beat. The spatial beats were analyzed by a least-squares fit of the data [Figs. 1(b) and 1(c)] using $I(\eta,H_0) - I(\eta,0)$ with $C(\eta,H_0) - C(\eta,0) = 0$. An adjustable phase of the trigometric function was permitted. The measured g value[7] and magnetic field were used. The parameters were the beat period $\bar{\eta}$ and the decay time constant τ, varied over the range of all plausible time constants.

In order to determine that our method is indeed valid, it was applied to transitions from the $3p'\,^2F_{7/2}^\circ$, $3d'\,^2G_{9/2}$, and $3p\,^2P_{3/2}^\circ$ levels of O II which have respectively short-lived, long-lived, and no apparent cascades. The lifetimes and cascading of these levels have been determined by the more elaborate analysis of BFS.[8] Table I compares the results of and the complications in the two methods. Our measurement technique at $H_0 = 0$ not only yields the BFS results but, with $H_0 \neq 0$, also permitted a third method, namely, a best-fit analysis of the decaying oscillating curve using $[A + B\cos(a\eta) + C\sin(a\eta)]\exp(-a\eta/2\omega_L\tau)$ with the constant background removed. When there was not any cascading, we obtained identical results by all three methods. In the presence of cascades, this third type of fit is not meaningful, as expected.

One should note our value of 6.8 nsec for the mean life of $3d'\,^2G_{9/2}$, a level which decays to $3p'\,^2F_{7/2}^\circ$. A standard BFS experiment on τ for

TABLE I. Comparison of lifetime measurements.

Charge state	Lower and upper level	Wavelength (Å)	Mean life (nsec) This method	Mean life (nsec) BFS	Calculated transition probability[a] (10^8 sec^{-1})	Other observed decay constants (nsec) BFS[b]	Other observed decay constants (nsec) This method	Wavelengths of cascades into levels from which transitions were observed (Å)
O II	$3s'\,^2D_{5/2}$–$3p'\,^2F_{7/2}{}^\circ$	4590.97	13.7 ± 0.4	14.0,[b] 12.8[b] 9.5,[c] 10.7[c] 12.0,[c] 14[c] 8.7[d]	1.11	4.0	None	4448, 4190, 4114
O II	$3p'\,^2F_{7/2}{}^\circ$–$3d'\,^2G_{9/2}$	4189.79	6.8 ± 0.2	6.1,[b] 5.05[b]	2.51	20.0	None	4253
O II	$3s\,^2P_{3/2}$–$3p\,^2P_{3/2}{}^\circ$	3973.26	6.8 ± 0.1	7.1,[b] 6.0[c]	1.27		None	

[a]See Ref. 12 [b]See Ref. 8. [c]See Ref. 10. [d]See Ref. 11.

the $3p'\,^2F_{7/2}{}^\circ$ level should then exhibit a cascade contribution with an effective mean life ≥ 6.8 nsec, since the $3p'\,^2F_{7/2}{}^\circ$ level might be repopulated by several higher terms. In fact, as Table I shows, the cascade τ in a standard BFS measurement was only 4.0 nsec. This illustrates the difficulty of determining the effective cascade lifetime in a standard experiment.

Our new technique is experimentally clearly superior to our beam-foil realization of Hanle-effect measurements on levels with mean lives $\gtrsim 3$ nsec.[1] For such longer-lived levels, appreciable integration times over the beam decay length are achieved only when the spectrometer slit is oriented parallel to the beam. Such a geometry is not amenable to spectrometer refocusing,[9] which provided the optical resolution necessary for the present measurements. Hence, a comparison with the Hanle effect was not feasible. A normal geometry with spectrometer slit perpendicular to the beam limits the spatial integration time for foil-populated levels, and makes Hanle-effect results prone to distance and velocity measurement uncertainties.[1]

We expect that the technique outlined here will serve as a useful and convenient adjunct to the usual BFS technique, to remove ambiguity and inaccuracy in cases where the presence of cascades introduces difficulties in analysis.

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Two-Photon Self-Induced Transparency of Different-Frequency Optical Short Pulses in Potassium*

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Low-loss, coherent two-photon propagation and pulse breakup with peak amplification are observed resulting from two-photon resonant self-induced transparency of different-frequency optical pulses interacting with a three-level system in potassium vapor. Theoretical analysis yielding new area equations for two different-frequency pulses with computer solutions agrees well with the observed results.

Novel propagation effects of coherent short light pulses have been studied in connection with a resonant interaction with a two-level system such as in ruby[1] and gaseous SF_6[2-4] and Rb.[5] Self-induced transparency (SIT) due to a one-photon resonant transition was first investigated by McCall and Hahn.[1] Recently, coherent two-photon propagation in which twice the propagating frequency is resonant with a two-level system has also been analyzed,[6,7] though the experimental study has not been reported yet. We wish to present here the first observation and theoretical analysis of a new type of coherent propagation of two different-frequency optical pulses causing a two-photon transition in a gaseous three-level system. The present model excludes two simultaneous transitions in double resonance.

The basic effect of coherent two-photon propagation can by analyzed in the limit of two different-frequency plane waves given as

$$E_i(z,t)=\epsilon_i(z,t)\cos[\omega_i t-k_i z+\varphi_i(z,t)] \quad (i=\lambda,\nu), \tag{1}$$

where the electric fields E_λ and E_ν induce the transitions between the energy levels 1-2 and 2-3 in a three-level system consisting of the ground state 1 and the two excited states 2 and 3, respectively. The frequencies ω_λ and ω_ν are assumed to be far off resonance from the eigenfrequencies Ω_λ and Ω_ν for each transition, although the sum of these frequencies $\omega_\lambda+\omega_\nu$ is equal to $\Omega_\lambda+\Omega_\nu$; i.e., resonant to the transition 1-3. Also, $\epsilon_i(z,t)$ and $\varphi_i(z,t)$ are considered slowly varying. A set of coupled equations of the time-dependent coefficients in the expansion of the wave function is obtained from the Schrödinger equation for the system. The solutions give the macroscopic induced polarizations, which act as sources in the self-consistent forms for the electric fields in accordance with Maxwell's equations. We can obtain from the reduced Maxwell's equations the area equations of different-frequency, two-photon propagation given by

$$\frac{d}{dz}\int_{-\infty}^{\infty}\epsilon_i^2\,dt=-\frac{\beta_i}{k}\left[1-\cos\left(k\int_{-\infty}^{\infty}\epsilon_\lambda\epsilon_\nu dt\right)\right], \tag{2}$$

where $\beta_i=4\pi\Omega_i N_0\mu_\lambda\mu_\nu/cn\hbar\Delta\omega$, $k=\mu_\lambda\mu_\nu/2\hbar^2\Delta\omega$, n is the refractive index of the medium, c is the light velocity in vacuum, $\Delta\omega=|\Omega_i-\omega_i|$ is the frequency deviation from the intermediate (virtual) state 2, μ_i is the matrix element for the electric dipole moment, and N_0 is the atomic density. Here it was assumed that the spectrum distribution function of the medium is uniform and the frequency shift from $\varphi_i(z,t)$ is negligible. The condition $T_2^*\ll\tau\ll T_2'$ is assumed for the actual pulse width τ. It is seen that Eq. (2) covers the single equation derived by Belenov and Poluektov[6]