

Multiphoton resonances and Bloch-Siegert shifts observed in a classical two-level system

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We report experimental observation of multiphoton resonances and Bloch-Siegert shifts in a strongly driven classical two-level system. The system is an optical ring resonator, and the levels are two orthogonal linear polarizations. The Rabi frequency and the Bloch-Siegert shift were measured for both the one- and three-photon cases. Satisfactory agreement was achieved with theory developed for a quantum two-level system. The experiment demonstrates that these coherence phenomena can be observed in a purely classical system.

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I. INTRODUCTION

We report the observation of multiphoton resonances and Bloch-Siegert shifts in a macroscopic classical two-level system. The interpretation of the effects observed in the experiment is based on the equivalence between the equation that describes the time development of this system and the Schrödinger equation for a driven two-level quantum system.

The driven two-level system has been extensively studied. In 1965, Shirley developed a formalism for a quantized atom in interaction with an oscillating classical field using Floquet theory [1]. Although his approach was semiclassical, he interpreted it as a classical approximation to the fully quantum treatment of the field. This interpretation is clear if it is recognized that Floquet states are essentially equivalent to dressed states in the limit of a large number of photons in the field. His formalism had no recourse on the rotating-wave approximation (RWA) and so is exact, even for strong fields. Within the context of this formalism he used perturbation theory to calculate the multiphoton Rabi oscillation frequency and Bloch-Siegert shift. In more recent years, the Bloch equation and the dressed-state formalism were used to calculate high-field coherence phenomena. Rabi oscillations, Bloch-Siegert shifts, multiphoton transitions, and the ac Stark effect have been extensively investigated for atoms and molecules in the optical regime [2], in microwave resonance [3,4], and in radio-frequency (rf) spectroscopy [5–8].

Although he developed his formalism for a quantum-mechanical two-level system, Shirley pointed out that “the Bloch-Siegert shift and all multiple quantum transitions derived above should appear in classical arguments,” and referred to Feynman, Vernon, and Hellwarth [9], who showed that the semiclassical Schrödinger equation for the two-level system is equivalent to the classical vector equation $d\mathbf{r}/dt = \boldsymbol{\omega} \times \mathbf{r}$, which is of course the basis of the Bloch vector approach [2,5].

It is also known that in the slowly varying envelope approximation (SVEA) the Maxwell equations for the classical electromagnetic field lead to an equation of the same

form as the Schrödinger equation [10–13]. This is the basis of studies into analogies in optics and quantum mechanics and makes it possible to create an optical system with two coupled states which is described by an equation identical to the Schrödinger equation for a quantum two-level system [14–16].

We have already reported the observation of some coherence phenomena in such an optical implementation of the driven two-level system; namely adiabatic following, Rabi oscillations, and Autler-Townes doublets [14,17]. Here we report the observation of multiphoton resonances and Bloch-Siegert shifts, together with a detailed discussion of the experimental method which was also used in previous experiments [14,18]. We show that the application of Shirley’s and Stenholm’s theories [1,5] for the quantum two-level system gives satisfactory agreement.

As already mentioned, we note that the theory of coherence phenomena has been developed extensively in the past. We do not intend to develop new theory in this paper; we show, rather, that an optical system can be made for which the same theory applies. The advantage of the optical system is that it is macroscopic, and consequently all parameters can be changed.

II. THE OPTICAL TWO-LEVEL SYSTEM

The classical two-level system used in this experiment is equivalent to one already reported [14–16,18] and is sketched in Fig. 1. In a planar optical ring resonator two electro-optic modulators are placed, one with its polarization axes along x and y (EOM1), and the other with its axes along $x + y$ and $x - y$ (EOM2). The modulators introduce a phase difference between the pair of light waves that are linearly polarized along their axes. This phase is proportional to the voltage applied to the modulator.

In this system we consider one longitudinal mode at an optical frequency ω_{opt} . For the empty resonator, the polarization modes x and y are degenerate. But if modulator EOM1 introduces a phase difference between the x - and y -polarization modes, this results in slightly different mode frequencies, which we denote $\omega_{\text{opt}} + W$ and $\omega_{\text{opt}} - W$, respectively. As the modes have principal fre-

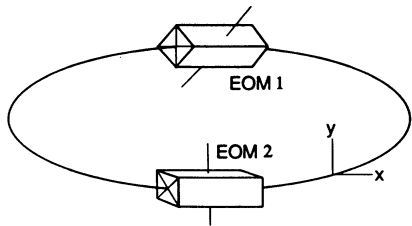


FIG. 1. Classical realization of a driven two-level system based on two orthogonal polarization modes in a ring resonator. EOM1 and EOM2 are electro-optic modulators, placed at an angle of 45° to each other. The polarization labels x and y are indicated.

quency ω_{opt} , the different frequency of the polarization modes can be expressed by letting the amplitude x of the x -polarized mode oscillate at frequency W , i.e., $x(t) \propto e^{-iWt}$, so that $dx(t)/dt = -iWx(t)$. The same argument for y leads to $y(t) \propto e^{+iWt}$ and $dy(t)/dt = +iWy(t)$. Using these amplitudes, the state of the system can be written as a complex column vector x, y , following the Jones vector formalism. The time derivative of this vector can now be written as

$$i \frac{d}{dt} \begin{bmatrix} x \\ y \end{bmatrix} = \begin{bmatrix} W & 0 \\ 0 & -W \end{bmatrix} \begin{bmatrix} x \\ y \end{bmatrix}. \quad (1)$$

This equation just describes the frequency deviation of the x and y polarization mode compared to the optical frequency ω_{opt} . When the frequency shifts of the $x + y$ and $x - y$ polarization modes due to EOM2, which we shall call $+S$ and $-S$, are included, one finds

$$i \frac{d}{dt} \begin{bmatrix} x \\ y \end{bmatrix} = \begin{bmatrix} W & S \\ S & W \end{bmatrix} \begin{bmatrix} x \\ y \end{bmatrix}. \quad (2)$$

The eigenvalues of the matrix are the frequencies of the eigenmodes, and are given by $\omega_{\pm} = \pm(W^2 + S^2)^{1/2}$. They form an avoided crossing with minimum level separation $\omega_0 = 2W$, as illustrated in Fig. 2.

Note that the same equation can also be derived for the

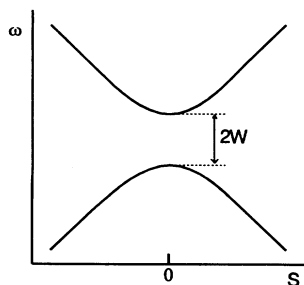


FIG. 2. Avoided crossing in the two-level system. The frequency of the modes is ω , the frequency splitting due to EOM1 is $2W$, and the detuning due to EOM2 is S .

amplitudes of the two propagation modes in the ring (clockwise and counterclockwise), using a partial reflector and rotation of the ring instead of electro-optic modulators. This model has been studied in mode-coupling theory for ring lasers [19,20], and has been used for earlier two-level experiments [14,17,21].

If an oscillating field with frequency ω and amplitude $2b$ [22] is applied to EOM2, so that the frequency shift S is $2b \cos(\omega t)$, then the time development of the system [Eq. (2)] becomes

$$i \frac{d}{dt} \begin{bmatrix} x \\ y \end{bmatrix} = \begin{bmatrix} W & 2b \cos(\omega t) \\ 2b \cos(\omega t) & -W \end{bmatrix} \begin{bmatrix} x \\ y \end{bmatrix}. \quad (3)$$

Note that in introducing the driving field, we choose an oscillating term for S , so that a counter-rotating term is present in the driving field.

If Eq. (3) is multiplied on both sides by \hbar , one finds exactly the Schrödinger equation for a quantum two-level system with level separation $\omega_0 = 2W$, driven by an oscillating interaction at frequency ω and amplitude $2b$. The matrix in Eq. (3) multiplied by \hbar is the Hamiltonian of the quantum-mechanical system. This is an example as to how the slowly varying envelope approximation in electromagnetism leads to equations of the same form as the Schrödinger equation. In the present case the SVEA corresponds to the assumption that the change of the intracavity field after a roundtrip is negligible (the intracavity field is described by position-independent amplitudes x and y). This is a reasonable assumption as typical frequencies in the experiment, such as the rf frequency of the driving field, are well below the free spectral range of the resonator.

Apparently the system can be described by the same equation as a quantum two-level system in interaction with a classical field. Therefore we can apply theory developed for the latter system. We will recall the results for the one- and three-photon Rabi oscillation frequency and Bloch-Siegert shift from Shirley [1], Stenholm [23], and Swain [24].

For small driving fields ($b/\omega \ll 1$) and near resonance ($\omega_0 \approx \omega$), it is reasonable for many cases of interest to make the rotating-wave approximation (RWA). The off-diagonal elements may be written as $2b \cos(\omega t) = be^{-i\omega t} + be^{+i\omega t}$. In the RWA, one of these terms (the counter-rotating term) is neglected, and then the problem is exactly solvable. The result is an oscillating amplitude for both levels, called Rabi oscillation, which occurs at frequency

$$\Omega_1 = 2b. \quad (4)$$

We shall call this the one-photon Rabi frequency, because a driving-field frequency equal to the transition frequency corresponds to a one-photon transition. Off-resonance the generalized Rabi frequency is given by

$$\Omega = (\Omega_1^2 + \Delta^2)^{1/2}, \quad (5)$$

where Δ is the detuning. In the experiment, however, the Rabi frequency is always measured on resonance, so that Eq. (4) applies.

If the counter-rotating term is taken into account, the

Rabi frequency is still given by Eq. (4), but the resonant transition frequency becomes smaller. This shift is called the Bloch-Siegert shift [25]. In first-order perturbation theory, valid for weak fields ($b/\omega \ll 1$), the new transition frequency becomes

$$\omega'_0 = \omega - \frac{b^2}{\omega}. \quad (6)$$

For larger field strengths, where perturbation theory is inappropriate, Stenholm gives a continued-fraction solution [23] to which Swain gives an analytical approximation [24]. The continued fraction solution has been tested, for example in magnetic resonance experiments [8].

When the transition frequency is an odd multiple of the driving frequency, multiphoton resonances occur. This results in a multiphoton Rabi oscillation. For a three-photon resonance the Rabi frequency, within third-order perturbation theory, is

$$\Omega_3 = \frac{b^3}{2\omega^2}, \quad (7)$$

while the three-photon transition frequency as a function of field strength, thus incorporating the three-photon Bloch-Siegert shift, is in second-order perturbation theory given by

$$\omega'_0 = 3\omega - \frac{3b^2}{2\omega}. \quad (8)$$

Stenholm gives a continued-fraction expression for the three-photon resonance frequency at high-field strength and again Swain gives an analytical approximation [23,24].

III. EXPERIMENTAL SETUP

The experimental setup used to measure the Rabi frequency and Bloch-Siegert shift consists of the classical two-level system as outlined in Sec. II, together with means to drive and probe the system, and is sketched in Fig. 3. The basis of the setup is a folded planar ring resonator with six curved high-reflecting dielectric mirrors. We used a ring resonator for convenience; in principle, the experiment could have been equally well performed with a linear (Fabry-Pérot) resonator. The free spectral

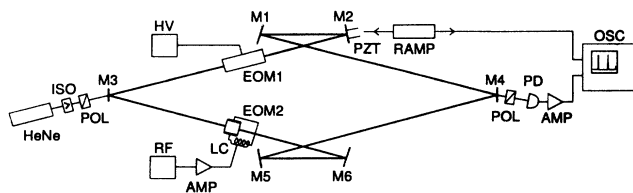


FIG. 3. The experimental setup. M1-6, high-reflecting dielectric mirrors; HeNe, frequency stabilized HeNe laser; ISO, optical isolator; POL, polarizer; EOM, electro-optic modulator; RF, radio-frequency signal generator; AMP, amplifier; LC, resonant LC circuit; HV, high-voltage source; PZT, piezomounted mirror; RAMP, ramp generator; PD, photodiode; OSC, oscilloscope.

range of the resonator, $\omega_{\text{FSR}} = 2\pi c/L$, was $2\pi \times 66.5$ MHz.

Two mirror pairs (M1,M2 and M5,M6) were used to optimize the beam waists inside the two electro-optic modulators placed inside this ring. One of these modulators, EOM1, was a high-voltage modulator with a half-wave voltage $V_{\lambda/2} = 4.0$ kV. It was placed with one of its polarization axes in the plane of the ring, and was connected to an adjustable dc voltage source. A voltage V on this modulator introduces a phase difference $\pi V/V_{\lambda/2}$ between the x - and y -polarized modes. Since a phase shift of 2π corresponds to a frequency ω_{FSR} , a voltage V on the modulator leads to a transition frequency

$$\omega_0 = \frac{V}{2V_{\lambda/2}} \omega_{\text{FSR}}. \quad (9)$$

The other modulator, EOM2, has a half-wave voltage $V_{\lambda/2} = 215$ V. It was placed with its axes at an angle of 45° with respect to EOM1. A coil was connected to this modulator, forming an LC circuit with the capacitance of the modulator. A rf signal at the resonance frequency of the circuit (4.35 MHz) was applied to a part of the coil. In this way the voltage amplitude from the rf source was transformed to a voltage across the modulator which was approximately 13 times higher. This enabled us to use low rf powers to create a large voltage across the modulator. The maximum voltage used in the experiment, using 1-W rf power, was about 130 V; higher values led to problems due to heating of the modulator crystal.

The field strength b can be calculated from the voltage across EOM2. When the ac voltage across EOM2 has an amplitude V , the maximum frequency difference between the $x+y$ and the $x-y$ polarization mode is $\omega_{\text{FSR}} V/2V_{\lambda/2}$. This should be equal to the sum of the off-diagonal elements in Eq. (3) which is at maximum $4b$, so that

$$b = \frac{1}{4} \frac{V}{2V_{\lambda/2}} \omega_{\text{FSR}}. \quad (10)$$

In order to detect the resonances, the spectrum of the ring was measured. This was done by injecting light from a fixed-frequency (633 nm) He-Ne laser, matched to the TEM₀₀ resonator mode, through one of the mirrors. The polarization of the injected light was chosen to be along one of the axes of EOM1. The light leaking through one of the other mirrors was analyzed with a polarizer, aligned parallel to the polarization of the injected light, and detected with a photodiode. In this way, only one state of polarization is injected with light, and its output subsequently detected.

We looked for resonances in this mode by scanning the roundtrip phase. This was done by scanning the length of the ring with a piezomounted mirror over a couple of wavelengths. While scanning, the photodiode signal was displayed on an oscilloscope. The scan frequency was small compared to all other frequencies occurring in the experiment, so that the detection is in effect stationary. The finesse of the resonator was about 50.

If no rf field is applied, the spectrum observed is as in Fig. 4(a). A resonance occurs at a particular ring length or roundtrip phase. The resonance of the other polariza-

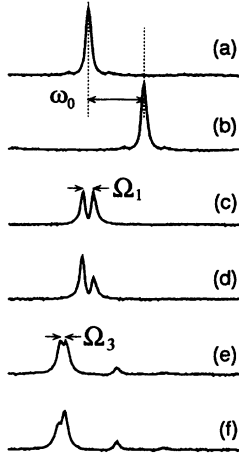


FIG. 4. Spectra of the ring resonator. (a) Spectrum of the x polarization with zero rf field. (b) Spectrum of the y polarization with zero rf field. (c) Doublet in the x polarization, caused by one-photon Rabi oscillation. (d) Same for small detuning from one-photon resonance. (e) Doublet due to three-photon Rabi oscillation. (f) Same for small detuning from three-photon resonance.

tion mode occurs at a different length of the ring because of the phase difference between the modes induced by EOM1 [Fig. 4(b)]. The distance between the resonances determines the transition frequency ω_0 , and could be changed by the voltage on EOM1.

If a rf field is applied to EOM2 and the transition frequency is tuned to the driving field frequency, a doublet appears in the spectrum [Fig. 4(c)]. We interpreted this as caused by the Rabi oscillation, and assumed the Rabi frequency to be equal to the splitting. When the transition frequency was changed slightly, the doublet became asymmetric [Fig. 4(d)], the splitting now corresponding to the generalized Rabi frequency [cf. Eq. (5)]. We interpreted resonance as occurring when the doublet is symmetric.

When the transition frequency was approximately three times the driving-field frequency, the one-photon resonance is far off-resonance, so that the doublet becomes highly asymmetric, consisting of a large and a small peak. When the driving-field amplitude was sufficiently high ($b/\omega \approx 1$), the larger peak is again split, due to the three-photon Rabi frequency, resulting in a spectrum like Fig. 4(e). By detuning from the three-photon resonance, this doublet could also be made asymmetric [Fig. 4(f)]. In Figs. 4(e) and 4(f) we have chosen by way of illustration the case of a small three-photon splitting, which is hardly resolved. This has the advantage that the assignment of the three peaks is directly clear. In the actual experiment discussed in Sec. IV the three-photon splitting was made much larger, so that it was fully resolved. The interpretation of the observed splittings in Figs. 4(c)–4(f) as Autler-Townes doublets [5] can be understood from the fact that the rf driven two-level system is probed with an optical field (Fig. 5).

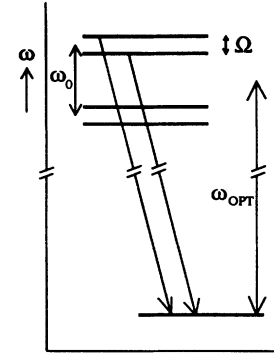


FIG. 5. The origin of the one-photon Autler-Townes doublet. The level splitting is ω_0 . When one of the levels split by the Rabi oscillation at frequency Ω , is probed to a third level which is an optical laser frequency ω_{OPT} away, two resonant frequencies occur.

IV. EXPERIMENTAL RESULTS

The one-photon Rabi frequency and resonant transition frequency were measured by first making the transition frequency ω_0 equal to the driving-field frequency ω . The transition frequency was then tuned to resonance by making the resulting doublet symmetric. On resonance, the Rabi frequency was determined by measuring the doublet splitting. Since the splittings were fully resolved, we performed no deconvolution.

In Fig. 6(a) the one-photon Rabi frequency is plotted as a function of the dimensionless driving field strength b/ω . The measured values are in accordance with the linear relationship predicted by Eq. (4), which is also shown in this figure. Since the amplification of the LC circuit on EOM2 was not known precisely, we fitted this value to get the correct slope, thus calibrating the field strength. The resulting value of the field strength was, however, within 10% of the estimated value. The errors in the measured values are mainly due to instabilities in the system, which caused the splitting observed in the spectrum to fluctuate.

The measured one-photon resonant transition frequency is plotted in Fig. 6(b) in a consistent way against b/ω using the same calibration. For high fields, it differs significantly from the perturbation expression [Eq. (6), drawn in the figure]. It is, as would be expected, in better agreement with the approximation to the continued-fraction solution given by Swain [24]. The large error bar for high driving-field amplitudes is due to the severe broadening of the resonance, as already pointed out by Cohen-Tannoudji, Dupont-Roc, and Fabre [6].

Subsequently, the three-photon Rabi frequency and resonant transition frequency were measured, by looking for resonance with $\omega_0 \approx 3\omega$. In Fig. 6(c) the three-photon Rabi frequency is plotted as a function of field strength, together with the curve expected [Eq. (7)]. The three-photon resonant transition frequency is plotted once again as a function of b/ω , in Fig. 6(d), together with the theoretical curves from perturbation theory [Eq. (8)] and

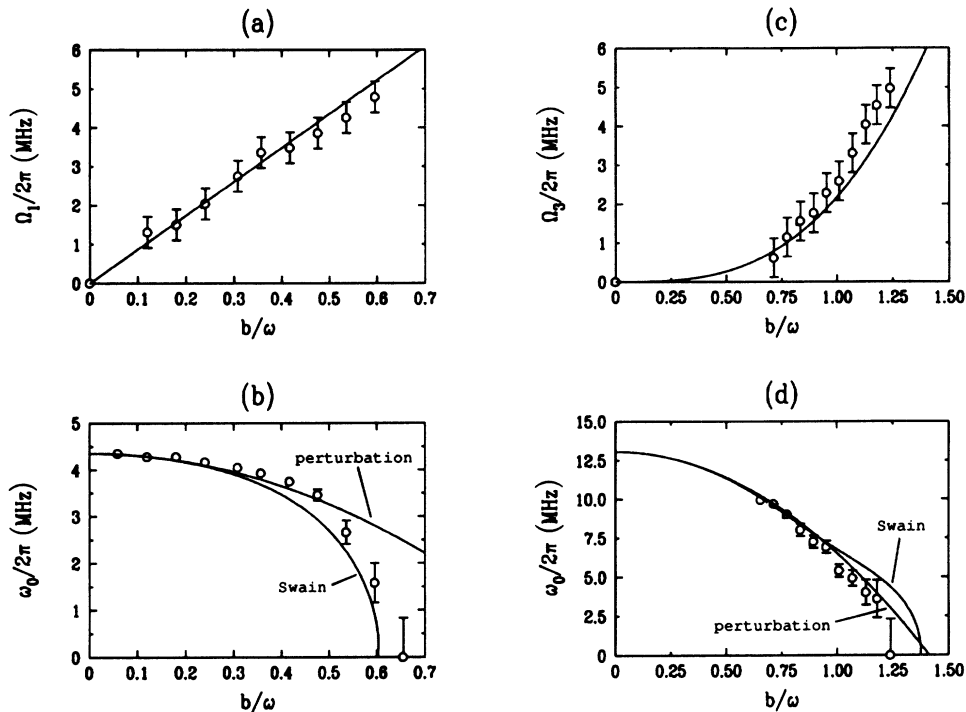


FIG. 6. The Rabi frequency and the resonant transition frequency as a function of the dimensionless driving field strength b/ω . The driving field oscillates at $\omega = 2\pi \times 4.35$ MHz. The points are experimentally observed; the lines are theoretical curves. (a) One-photon Rabi frequency. (b) One-photon resonant transition frequency. (c) Three-photon Rabi frequency. (d) Three-photon resonant transition frequency.

Swain [24]. Both figures show satisfactory agreement with theory. It is somewhat surprising that for the three-photon Bloch-Siegert shift the perturbative expression is rather close to the analytical approximation to the continued-fraction solution.

Direct comparison may readily be made between the Shirley model and the rather more familiar semiclassical picture appropriate to multiphoton absorption from lasers [26]. The intermediate Floquet states have their counterpart in nonresonant virtual intermediate states and multiphoton resonance and level shifts are again found. In each formulation there are restraints imposed by symmetry. A pair of levels connected by a one-photon electric dipole transition will only show odd-multiple-quantum transitions, while levels of the same parity only demonstrate even-multiple-quantum transitions. Similar behavior is found for the classical atom where due to the symmetry of the system only the odd-photon resonances occur. However, experimental evidence was found of two-photon resonances when the prevailing symmetry was broken by applying a dc offset to the rf modulator EOM2.

V. CONNECTION WITH OTHER TWO-LEVEL SYSTEMS

A natural question arises as to how this two-level system relates to other two-level systems. The response of

our system depends on the electric field inside EOM2, and as a result of an oscillating electric field a Rabi oscillation with a certain frequency is observed. We can compare this with the response of a system with an electric dipole moment, such as the atomic [26] or molecular case [7], by calculating the “dipole moment” of the ring. For a system with an electric dipole moment M_1 , the Rabi frequency, resulting from an oscillating electric field with amplitude \mathcal{E} , is [2]

$$\Omega_1 = \frac{2M_1\mathcal{E}}{\hbar}. \quad (11)$$

In our system, the rf voltage on EOM2 is applied to a crystal whose electrodes are approximately 5.0-mm apart. A rf amplitude of 67 V, which corresponds to an electric-field strength of 13.4 kV/m, gave a Rabi frequency of 5.0 MHz. With Eq. (11), this leads to a dipole moment of 0.037 Debye [27].

For a resonant frequency of ≈ 4 MHz the dipole moment of 0.037 Debye yields an Einstein A coefficient $\approx 1.4 \times 10^{-21} \text{ s}^{-1}$ and a lifetime of $7 \times 10^{20} \text{ s}$ for the upper level [28]. Consequently spontaneous decay would not be expected any more than it would be in rf spectroscopy of atoms and molecules. What is important, however, is the Einstein B -coefficient of $\approx 8 \times 10^{20} \text{ cm}^3 \text{ J}^{-1} \text{ s}^{-1}$,

which allows the classical or rf atom to be driven by a resonant or near-resonant field.

As a typical atomic or molecular dipole moment is $ea_0 = 2.5$ Debye [27] (where e is the elementary charge and a_0 is the Bohr radius of an atom), this means that if the same electric field as inside EOM2 were applied to an atom, the Rabi frequency would be approximately 70 times larger. The fact that the frequency of the driving field in our classical system can be chosen low, enabled us to make b/ω of order unity. This is large enough to see the high-field effects, and corresponds roughly to the same b/ω value involved in the rf spectroscopy experiments [8]. In our system an even larger value of b/ω could be made, with the same field, by using a lower driving-field frequency or, as can be seen from Eq. (10), by use of either a modulator with a lower half-wave voltage or a resonator with a larger free spectral range, i.e., by using a smaller resonator. The limiting factor here is the finesse of the resonator, which must be large enough to be able to resolve the Rabi splitting.

It should be noted that our system does also allow an interpretation in terms of magnetic dipole resonance (spin- $\frac{1}{2}$ system). This point of view, and its connection with the electric dipole resonance interpretation is discussed in Ref. [15].

VI. CONCLUSION

We have demonstrated multiphoton transitions together with their Bloch-Siegert shifts in a classical macroscopic optical system. One electro-optic modulator was used to create two polarization eigenmodes with a certain frequency difference, and another to couple them with an oscillating field. The measured Rabi frequencies and transition frequencies for the one- and three-photon resonances agreed reasonably well with the theory for quantum two-level systems. It should be stressed that no free parameters were available to fit the theory to the experiment other than a minor "correction" to the field strength. The experiment clearly demonstrates that these effects can be observed in a purely classical system.

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- $$\Delta_1(x) = \frac{15x^4}{2} \left[16 + 4x^2 - \frac{9x^4}{4} - \frac{10x^2/3}{1 - 7x^2/60} \right]^{-1}$$
- (private communication with S. Swain).
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