PHYSICAL REVIEW LETTERS

VOLUME 18 | 1 MAY 1967 | NUMBER 18

COHERENT DETECTION OF A NEW TYPE OF OPTICAL DOUBLE-QUANTUM TRANSITION*

W. Happer and B. S. Mathur

Columbia Radiation Laboratory, Columbia University, New York, New York

(Received 27 March 1967)

We wish to report on experimental observations of a new type of optical double-quantum transition[1] in which an atom passes from one Zeeman sublevel to another by virtual absorption and successive re-emission of optical photons. A remarkable property of these transitions is that the energies of the optical photons involved are on the order of 10^{10} times larger than the energy difference between the initial and final states of the Zeeman transition. Spontaneous two-photon transitions have been observed recently[2] in the decay of the $2S_{1/2}$ state of He^+. Stimulated two-photon transitions between discrete atomic states[3] and multiple-quantum photoionization[4] have also been observed. In previous work it has been necessary to use high-intensity laser beams to induce optical multiple-quantum transitions. The present experiment uses the rather weak light fluxes which are produced by ordinary resonance lamps. The double-quantum transitions are still observable because of three factors: The intermediate states in the double-quantum transition are nearly resonant; the initial and final states in the transition are different Zeeman sublevels of a long-lived optically pumped atom; and a sensitive coherent detection system is used in the spectrometer. The double-quantum transitions studied in this work are quantitatively related to the light shifts[5] caused by virtual transitions in optically pumped atoms. The spectrometer used in this experiment is sensitive enough to detect light shifts as small as 1 part in 10^4 of the natural linewidth of the Zeeman resonance. This experiment is also closely related to the Raman effect.[6] In the Raman effect one observes the sidebands introduced onto light following optical double-quantum transitions between different initial and final states. In this experiment the occurrence of an inverse Raman process is detected by observing the reorientation of the atoms.

The basic concepts of the experiment are illustrated in Fig 1. A beam of monochromatic light is incident on an atom which is in the

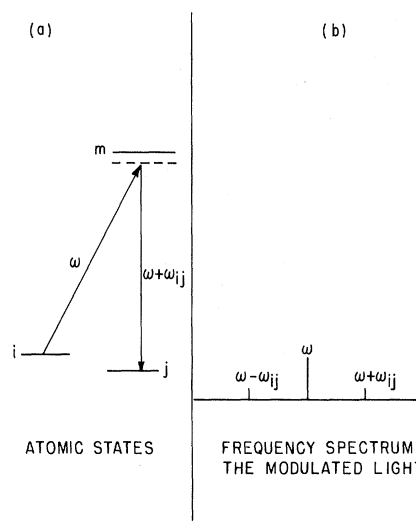

FIG. 1. Double-quantum transitions between two Zeeman levels caused by optical photons.

sublevel i of its ground state. The frequency ω of the light is several Doppler widths removed from one of the optical absorption frequencies of the atom. When the light is amplitude modulated at the ground-state transition frequency ω_{ij}, one finds that the atom makes a transition from the level i to the level j of the ground state. The mechanism for the transition is the following: The amplitude-modulated light contains a carrier frequency ω and two sideband frequencies $\omega \pm \omega_{ij}$. The atom absorbs a photon at the carrier frequency ω and is excited to the virtual state m. Then one of the sidebands of the modulated light stimulates the atom to re-emit a photon of frequency $\omega + \omega_{ij}$ and fall down to the state j. The frequencies are such that energy is conserved in the process, and the polarization of the incident light must be such that angular momentum is also conserved.

The actual experiment was performed with optically pumped rubidium vapor. Since alkali atoms have rather complicated hyperfine structure in the ground state and in the excited states, there are many possible double-quantum transitions of the type illustrated in Fig. 1 involving different intermediate states m and different combinations of sideband and carrier frequencies. The amplitudes of all of these double-quantum processes add coherently to produce the final transition amplitude. However, it can be shown that to a high degree of approximation, the net effect of all double-quantum transitions within the Zeeman multiplet F of the ground state can be represented by an effective Hamiltonian of the form $\delta\vec{H}\cdot\vec{\mu}$, where $\vec{\mu}$ is the dipole moment operator of the atom and $\delta\vec{H}$ is a small fictitious magnetic field, defined by the expression

$$\delta\vec{H} = \frac{\lambda^2 u}{4\pi g_J \mu_0}\,\frac{e^2}{mc^2} f_J \,\frac{\omega}{\omega-\omega_F} \times\left[\frac{11-4J(J+1)}{4}\right]\left(\frac{\hat{e}\times\hat{e}^*}{i}\right). \qquad (1)$$

Here λ is the wavelength of the exciting light, and $\omega = 2\pi c/\lambda$ is the corresponding angular frequency. The absorption oscillator strength to the ^2P_J doublet is f_J; ω_F is the average frequency separation between all of the hfs components of the ^2P_J excited state and the hfs multiplet F of the $^2S_{1/2}$ ground state. The electronic angular momentum of the ^2P_J excited state is J. The gyromagnetic ratio of the $^2S_{1/2}$ ground state is g_J, μ_0 is the Bohr magneton, and e^2/mc^2 is the classical electron radius. The energy density of the light beam averaged over a few optical cycles is u, and $\hat{e}$ is the complex polarization vector of the exciting light. Although the introduction of a fictitious magnetic field may seem to obscure the physics of the situation, it is actually a great conceptual help in understanding the design of this experiment. As far as the Zeeman coherence of the atom is concerned, the effects of the fictitious magnetic field are indistinguishable from those of a real magnetic field, and intuition gained from magnetic resonance work can be applied directly to effects caused by optical double-quantum transitions. In particular, this experiment can be described as a Zeeman resonance experiment with a fictitious oscillating magnetic field. The use of Eq. (1) also allows one to make quantitative estimates of the signal strengths which can be expected.

The experimental apparatus used to detect the optical double-quantum transitions is sketched in Fig. 2. Rb^{87} vapor in a 100-ml Pyrex flask was optically pumped by Rb^{87} resonance radiation. The flask was placed in the center of a solenoid 40 in. long and 12 in. in diameter. The solenoid was surrounded by two concentric cylindrical Mumetal shields. With this arrangement, Zeeman resonance linewidths were on the order of 40 Hz as monitored by observing the absorption of the pumping light. Off-resonant light was obtained by passing Rb^{85} light through a heated Rb^{87} filter cell, thus leaving only $F = 2$ hfs component of the Rb^{85} resonance

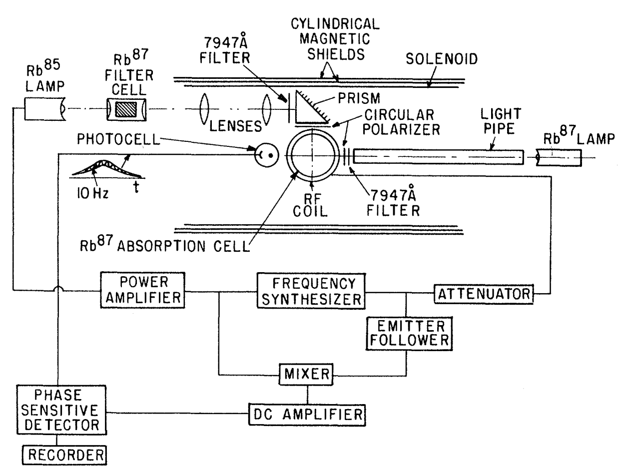

FIG. 2. The spectrometer for coherent detection of optical double-quantum transition.

light.[7] After passage through lenses, a prism, a 7947-Å interference filter, and a circular polarizer, about 10^{14} photons were incident on the optically pumped vapor. The equivalent transverse magnetic field δH [see Eq. (1)] was on the order of 10^{-7} G. The Rb^{85} light was 100% square-wave modulated with a 10 000-Hz signal from a frequency synthesizer. If δH were several orders of magnitude larger, it could be used to induce Zeeman resonances directly. However, for very small effective magnetic fields one can obtain a considerable enhancement in signal power (on the order of 10^5 in this experiment) by simultaneously applying a real, coherent, rf magnetic field. Thus, a real 9990-Hz transverse rf field was also applied to the absorption flask. The strength of 9990-Hz rf was attenuated empirically until the Zeeman resonance was half saturated. When the static magnetic field was swept through the resonance, a typical increase in the absorption of the vapor was observed as is indicated in Fig. 2. However, the fictitious magnetic field due to the optical double-quantum transitions alternately increased and decreased the transition probability so that a 10-Hz ripple was present on top of the usual resonance signal. The 10-Hz ripple was detected with a phase-sensitive amplifier and recorded on chart paper. A typical recorder trace is shown in Fig. 3. The 10-Hz signal disappeared when the Rb^{85} light was blocked or when the 10 000-Hz modulation was removed from the lamp. It also disappeared when the circular polarizer was removed from the beam, when the 9990-Hz rf was absent, or when the Rb^{87} pumping light was turned off. The signal strength was essentially independent of the temperature of the Rb^{87} filter cell once the cell was hot enough to absorb all of the $F=3$ hfs component of the Rb^{85} light. The signal amplitude and the signal-to-noise ratios were in agreement with order of magnitude theoretical estimates. We conclude that the observed signals do indeed represent optical double-quantum transitions between the Zeeman sublevels of the ground state.

Although these measurements were carried out with light, analogous experiments could clearly be performed with microwaves replacing the light and different hfs states of the atomic ground state replacing the excited state. A spectrometer analogous to the one sketched in Fig. 2 could then be used to carry out extremely precise measurements of microwave Bloch-Siegert[8] shifts due to neighboring hfs multiplets, or, conversely, one could make extremely precise absolute measurements of microwave magnetic field amplitudes. It is also clear that analogous effects should occur if the light is amplitude modulated at the hfs frequencies of the atomic ground state.[9] Thus an important application of this technique would be in the design of sensitive detectors of hfs-modulated alkali resonance light. In such a system optically pumped atoms would mix the light modulation frequency with a reference microwave frequency. Since the difference frequency would appear as a low-frequency amplitude modulation of the optical-pumping light beam, the only source of noise would be the shot noise of the light beam.

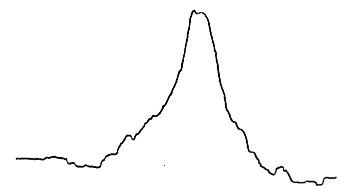

FIG. 3. A recorder trace of an optical double-quantum transition probability versus magnetic field. The integration time was 10 sec.

The authors would like to thank Mr. Richard Brignoli and Mr. Henry Tang for their assistance in this experiment. They also gratefully acknowledge the interest and encouragement of Professor R. Novick and Professor A. Lurio.

*This work was supported wholly by the Joint Services Electronics Program (U. S. Army, U. S. Navy, and U. S. Air Force) under Contract No. DA-28-043 AMC-00099(E).

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