Experimental Evidence of Two-Photon Transition without Doppler Broadening

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Experiments on the $3S$–$5S$ two-photon transition in sodium give evidence that Doppler broadening is eliminated if the atom absorbs two photons propagating in opposite directions. The proof is given by the comparison of the two-photon absorption line shape in traveling and standing waves.

Doppler-broadening elimination in multiphoton transitions has been theoretically studied in a recent paper.[1] In the present note, we report the first experimental demonstration of this theory.

We first summarize the principle of the method.[2] Consider an atom moving at a velocity $\vec{v}$, in a standing wave of circular frequency ω. In its rest frame, this atom interacts with two oppositely traveling waves of frequencies $\omega(1-v_x/c)$ and $\omega(1+v_x/c)$, respectively. We suppose that the atom can reach an excited state e (energy $\hbar\omega_e$) by absorbing two photons in the ground state g (energy $\hbar\omega_g$). At resonance, the following condition is fulfilled:

$$\hbar\omega_e-\hbar\omega_g=\hbar\omega(1-v_x/c)+\hbar\omega(1+v_x/c)=2\hbar\omega. \quad (1)$$

The terms depending on the velocity of the atom disappear, indicating that, at resonance, all the atoms, irrespective of their velocities, can absorb two photons. Theoretically the width of this resonance is of the same order of magnitude as the natural linewidth. If ω does not fulfill the resonant condition (1), but is still close to it, the atoms cannot absorb two photons propagating in opposite directions, although some atoms of definite velocity v_x can absorb two photons propagating in the same direction. The signal due to these two-photon transitions is of course much smaller than the one due to the oppositely traveling photons. We have tested the theory by doing experiments with sodium.

Figure 1(a) shows a simplified energy diagram of sodium. An atom can undergo a transition from the $3S$ ground state to the $5S$ excited state by absorbing two photons whose wavelengths are 6022.3 Å. We detect the atoms in the $5S$ state by collecting the photons spontaneously emitted from this state to the $3P_{1/2}$ and $3P_{3/2}$ levels at 6154 and 6160 Å.

The problem is in fact slightly complicated by the hyperfine structure of the $3S$ and $5S$ levels. In a transition from a state $L=0$ to a state $L=0$ (L is the orbital angular momentum), the selection rules are $\Delta F=0$ and $\Delta m_F=0$.[1] The signal will therefore be a superposition of the signals resulting from the transitions $F=1\rightarrow F'=1$ and $F=2\rightarrow F'=2$ [Fig. 1(b)]. The hyperfine structure of the ground state $3S$ is well known to be 1771 MHz. On the other hand, the hyperfine structure of the $5S$ level has never been measured. It can be estimated, from calculations based on the Fermi-Segre formula, to be about 155 MHz. By using these two values, one finds that the energy difference between the transitions $F=1\rightarrow F'=1$ and $F=2\rightarrow F'=2$ is equal to 1616 MHz. Experimentally, we monitor the strength of the absorption as a function of ω, the frequency corresponding to half the energy needed for the transition. We therefore expect to observe two peaks separated by about 800 MHz.

We use a flashlamp-pumped rhodamine 6G dye laser similar to the one described by Gale.[3] We have not yet succeeded in obtaining a monomode oscillation of the laser. During the pulse we ob-

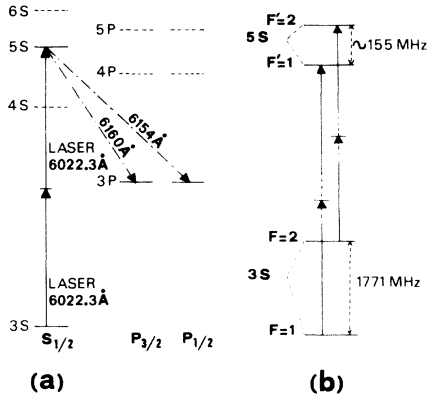

FIG. 1. Energy diagram of Na. (a) Levels which are involved in the two-photon transition. (b) Hyperfine components of the two-photon transition.

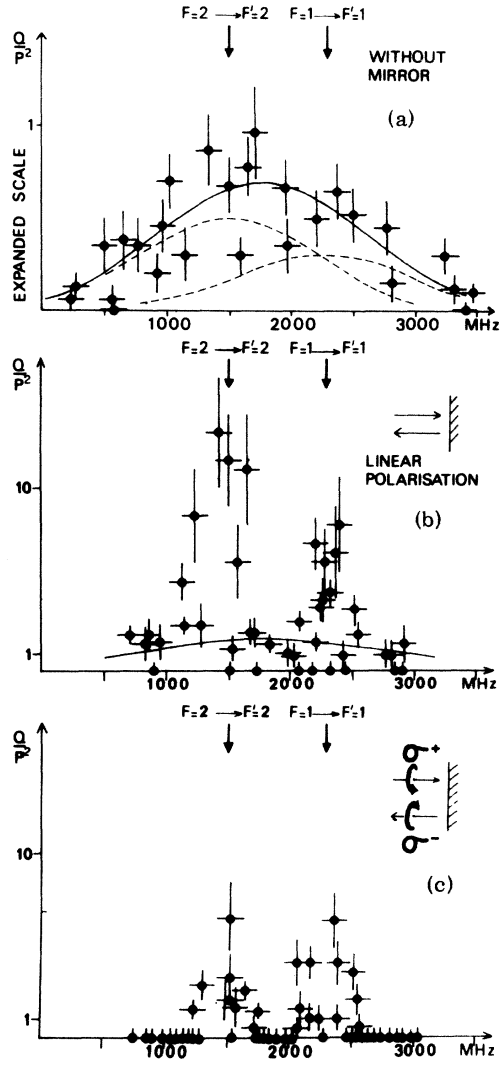

FIG. 2. Experimental results: The signal Q divided by the square of the laser power P is plotted versus the frequency of the laser; the origin of the X axis is arbitrary. (a) Two-photon transition with only one traveling wave. (b) Two-photon transition in a standing wave. (c) Two-photon transition with two traveling waves of opposite directions and different polarizations (σ^+ and σ^-).

serve three or four adjacent longitudinal modes, for which the distance between two adjacent modes is about 240 MHz. The mean frequency of the laser is measured by a Michelson interferometer which enables us to determine the relative values of the light frequency as indicated on the X axis of Fig. 2.

The peak power of the laser is about a few hundred watts. We measure the relative values of the power P of the laser for each pulse with a photodiode. A monochromator selects the photons re-emitted from the sodium vapor cell for wavelengths between 6152 and 6162 Å. The photons are detected by a photomultiplier. We measure its electric charge Q integrated over the pulse of duration 300 nsec. In Fig. 2, we plot on the Y axis Q/P^2 (in arbitrary units) since the two-photon transition probability is proportional to P^2.

The dispersion of the experimental points can be explained by the substantial nonreproducibility of the laser oscillation. Furthermore there are not many photoelectrons emitted during a pulse. In the best cases we have a few hundred, while in less favorable cases we have fewer than ten photoelectrons and the statistical fluctuations become important. The temperature of the sodium cell is about 220°C.

The experimental results obtained under the above mentioned conditions are presented in the three diagrams of Fig. 2. Figure 2(a) shows a preliminary experiment: The atoms interact with only one traveling wave *(without mirror)*. In this case, the theoretical curve is the sum of the Doppler lines corresponding to the two hyperfine transitions $F=2 \rightarrow F'=2$ and $F=1 \rightarrow F'=1$ [these Doppler lines are the dashed lines in Fig. 2(a), the sum of these Doppler lines is a solid line]. The experimental points are spread over a range of 2000 MHz with a similar intensity. The intensity of the maximum is much smaller than the ones of Figs. 2(b) and 2(c), which show resonances in the absence of the Doppler effect [the vertical scale of Fig. 2(a) is expanded by a factor of 10].

Figure 2(b) shows the experimental points obtained when the laser beam is *reflected back in the cell.* The laser beam is *linearly polarized.* In this case the atoms can either absorb two photons of the same wave, or two photons propagating in opposite directions. The Doppler line due to absorption of photons of the same wave is printed on Fig. 2(b), the intensity of this line is much smaller than that of the two narrow peaks which correspond to the two hyperfine transitions without Doppler broadening. The present uncertainty in the laser frequency prevents us from giving any significance to the experimental widths of the peaks, or assigning a precise value to the hyperfine structure of the 5S level.

Figure 2(c) shows the experimental points when *one traveling wave is polarized* σ^+ *and the other is polarized* σ^-. This is accomplished by placing one quarter-wave plate before the sodium cell, and another one between the cell and the mirror. In this case, the atom must absorb one photon of

each wave since the selection rule $\Delta m_F = 0$ forbids the absorption of two σ^+ or two σ^- photons.[1] One sees that the experimental signal is equal to zero outside the frequency range of the two peaks. The Doppler background disappears by using this polarization configuration. We have performed another test by removing the mirror and observing that the signal disappears since the atom cannot absorb two photons of a σ^+ traveling wave.

These experiments present evidence that we can eliminate Doppler broadening by two-photon transitions. Nevertheless, because of the relatively large spectral bandwidth of our laser, we are not yet able to attain the ultimate precision inherent in this method.

An improvement of our laser stability will permit a comparison between this method and other methods of spectroscopy without Doppler broadening.

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Observation of Two-Photon Absorption without Doppler Broadening on the 3S-5S Transition in Sodium Vapor*

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The $3S$-$5S$ transition of Na atoms is induced by absorption of two photons, one each from two circularly polarized beams traveling in opposite directions through the vapor. The beams are derived from a tunable, narrow-band pulsed dye laser. Doppler broadening is absent and the hyperfine structure is resolved. The hyperfine interaction constant A_{5S} in the $5S$ state of the Na^{23} atom is determined to be 101 ± 15 MHz.

Several authors[1-3] have recently called attention to the predicted absence of Doppler broadening in two-photon absorption processes, if the two photons have equal and opposite momenta. In this note we report the experimental observation of this effect for the $3S$-$5S$ transition in Na vapor. The novel technique permits high-resolution optical spectroscopy of many highly excited states, with energies up to twice the laser frequency, in atomic and molecular spectra. It complements and extends the high-resolution "Lamb-dip" techniques, based on saturation of one-photon transitions.[4]

Two-photon absorption may be described phenomenologically by the imaginary part of a nonlinear susceptibility, $i\chi^{(3)\prime\prime}$. This nonlinearity is relatively large in the alkali-metal vapors, and a variety of other related nonlinear optical processes, such as third-harmonic generation and frequency mixing, have recently been reported.[5-7] The thrust of those investigations was directed toward ultraviolet generation, infrared detection, or other nonlinear optical devices. This note links the nonlinear optical properties of alkali vapors to high-resolution spectroscopy. Since the precise value of the two-photon absorption cross section is not of primary present interest, a rough evaluation of the expected magnitude of the effect should suffice. Tabulated values of the matrix elements[7] and standard expressions for the two-photon absorption cross section indicated that under the experimental conditions described below about $1:10^7$ of the incident beam intensity would be absorbed by two-photon processes in the cell. The occurrence of these events is most conveniently detected by subsequent fluorescence radiation from the excited Na atoms. A cross section of this magnitude should provide an ample signal-to-noise ratio. This type of experiment has become feasible because of the availability of pulsed high-power, narrow-band, tunable dye lasers.

If the laser frequency is tuned so that $2\hbar\omega$ corresponds to the separation of two energy levels with the same parity, each atom in the gas, regardless of its thermal velocity, is at resonance for the two-photon absorption process in which one photon is taken from each of two counter-