Resonant Two-Photon Spectroscopy in a Fast Accelerated Atomic Beam

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Two-photon spectroscopy has been performed in a fast, accelerated atomic beam, with a resolution limited only by the natural linewidth of the upper excited level. The main feature of the experiment is the "creation" of a completely harmonic three-level atom, by using the relativistic transformation between the laboratory and atom rest frame. Thus resonant two-photon absorption, with a strength comparable to electric-dipole transitions, takes place. The systematic effects of importance in spectroscopy are investigated.

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Nonlinear, Doppler-free spectroscopy has been developed into a versatile tool for the study of atoms and molecules. In high-resolution spectroscopy, the two most used techniques are saturated absorption[1] and two-photon absorption,[2] both of which eliminate the Doppler effect normally associated with the spectroscopy of moving absorbers. High spectral resolution can also be obtained in collinear fast-beam laser spectroscopy[3,4] as a result of the velocity-bunching effect, which cools the ions or atoms along the direction of propagation. Because of variations of the beam velocity, the residual Doppler broadening can be eliminated only by using the nonlinear techniques, and so far, saturated absorption has been applied successfully to fast-accelerated atom/ion beams.[5,6]

It will be demonstrated in this work that high-resolution, two-photon spectroscopy can be performed on a fast, metastable atomic beam. The main features are the creation of a completely harmonic three-level atom, by using the relativistic transformation between the laboratory and the atom rest frames, followed by the absorption of two opposite-running collinear laser fields (of the same wavelength) by the fast-moving atoms. The effects of a resonant intermediate state have been studied experimentally by Björkholm and Liao,[2] who used two different laser fields, whereas Salomaa and Stenholm[7] and Brewer and Hahn[8] have treated this totally resonant case theoretically.

In the experiment, shown in Fig. 1, a $^{20}Ne^{+}$-ion beam is charge exchanged in sodium vapor, producing large amounts of the $3s[\frac{3}{2}]_2$ metastable level in Ne I. The velocity of this metastable level can be varied by applying a postacceleration voltage V_p to the exchange cell. After the exchange, the Ne* atoms interact collinearly with two counter-propagating laser fields. The scattered light is monitored by a spectrometer and a photomultiplier. The exciting-laser wavelength is measured with use of a real-time fringe-counting interferometer[9] (λ meter) absolutely calibrated to within ± 50 MHz and with a relative reproducibility better than ± 5 MHz.

The velocity of the fast Ne* I metastable beam is determined by using the λ meter and observing a single-photon transition, with the laser beam co-propagating and counter-propagating, respectively, along the fast-atom beam. Thus we have

$$\sigma_{L\pm}=\sigma_0\frac{[1-(v/c)^2]^{1/2}}{1\pm(v/c)}, \quad (1)$$

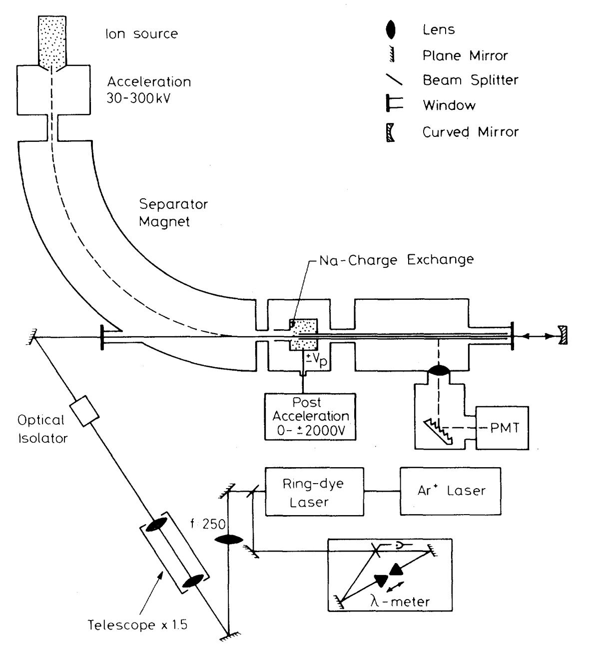

FIG. 1. Experimental apparatus consisting of a 300-kV isotope separator equipped with universal ion source and postacceleration in connection with the charge exchange. Also shown is the stabilized dye laser and λ meter.

which completely determines the velocity v of the atomic beam and the wave number σ_0, with the laser wave numbers σ_{L+} and σ_{L-} measured for opposite-running laser beams, respectively. The velocity v is determined to within 5×10^{-6}, by using this procedure. The wave number σ_0 for the $3s[\frac{3}{2}]_2$-$3p'[\frac{1}{2}]_1$ transition is found to be 16 996.615(1) cm^{-1}, in full agreement with Hall and Lee[9] and close to the value 16 996.612 cm^{-1} recommended by Kaufman and Edlén.[10]

The basic level scheme studied is shown in Fig. 2. To Doppler shift $3p'[\frac{3}{2}]_2$ into exact resonance between $3s[\frac{3}{2}]_2$ and $4d'[\frac{5}{2}]_3$, a beam velocity v must be selected as

$$\sigma(3p',3s)=\sigma_0\frac{1+(v/c)}{[1-(v/c)^2]^{1/2}},\qquad \sigma(4d',3p')=\sigma_0\frac{1-(v/c)}{[1-(v/c)^2]^{1/2}},\tag{2}$$

where $\sigma(\alpha,\beta)$ is the transition energy for the $\alpha\rightarrow\beta$ transition and σ_0 is the laser wave number measured in the laboratory frame. Using $\sigma(\alpha,\beta)$ from Moore,[11] we find the desired resonance at a beam energy of 119.141 kV and a laser number of $\sigma_0=16\,876.921$ cm^{-1}. In Fig. 3 is shown a laser-frequency scan around σ_0. A sharp Doppler-free, two-photon resonance with a width of $\simeq$4.7 MHz is observed. This width has to be compared to a first-order Doppler shift of $\simeq$ 2 THz, a second-order Doppler shift of $\simeq$ 3 GHz, and a velocity spread of the fast atoms corresponding to $\simeq$ 150 MHz ($\pm$ 10 V). Also shown in Fig. 3 is the intensity of the two-photon resonance as a function of beam energy. The resonant enhancement is clearly seen when tuning the $3p'[\frac{3}{2}]_2$ level through complete resonance.

These findings, along with several remarkable features, can be interpreted following Salomaa and Stenholm,[7] who suggested the use of the first-order Doppler shift to acheive the resonant enhancement from an intermediate level.

First, we find no ac Stark shifts of the resonance; second, only small power-broadening effects are observed; and third, a small broadening

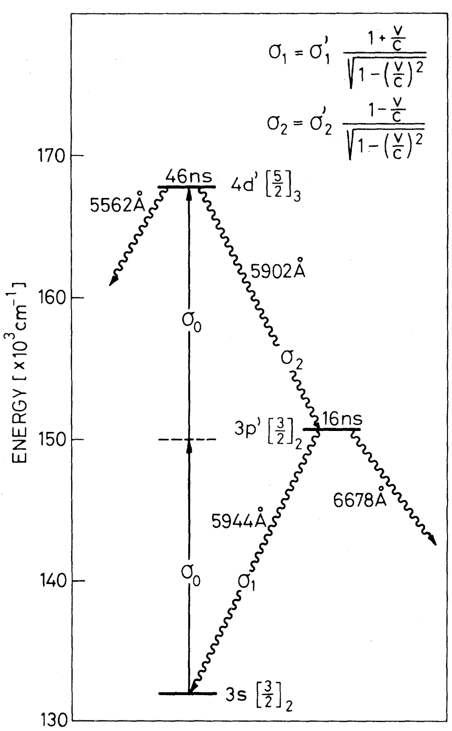

FIG. 2. Reduced energy-level diagram of Ne I showing the harmonic three-level system studied. Also shown is the relativistic transformation, which will locate the two-photon resonance at (v_0, σ_0), where $\sigma_0=\sigma_1'=\sigma_2'$.

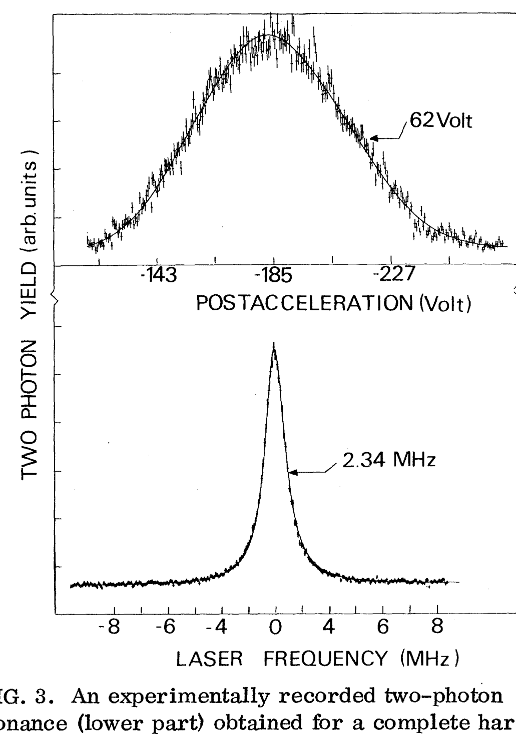

FIG. 3. An experimentally recorded two-photon resonance (lower part) obtained for a complete harmonic three-level atom (V_p = 186 V) in the decay $4d'$-$3p$ at 5562 Å. The resolution of $\simeq$ 4.7 MHz represents the highest one obtained in fast-beam, optical spectroscopy. Also shown is the resonant enhancement (upper part). The laser frequency is fixed at σ_0 and the apparent energy of the $3p'$ level is changed by varying the post-acceleration. A small adjustment of σ_0, due to the second-order Doppler shift is performed.

of $\delta\sigma \simeq \sigma_0 v\,\delta v/c^2 \simeq 1$ MHz is present as a result of the second-order Doppler effect.

Salomaa and Stenholm[7] have considered the three-level system shown in Fig. 2, with a strong field Ω_1 and a weak field Ω_2. On the assumption that $ku \gg \gamma_\alpha, \gamma_\beta$, where u is the spread in beam velocity, they found the population of the upper $4d'$ level to be given by[7]

$$\mathcal{L} \propto \exp\left(-\frac{\Delta_1^2}{(ku)^2}\right) \operatorname{Im}\left(\frac{F_-}{\Delta_0 - i\Gamma_-} + \frac{F_+}{\Delta_0 - i\Gamma_+}\right),$$

where $\Delta_1 = \omega_{3p',3s} - \Omega_1$, $\Gamma_\pm$ are the widths of the resonances with strength $F_\pm$, and $\Delta_0 = \omega_{4d',3s} - \Omega_1 - \Omega_2$. Evaluating $F_\pm$ with our relaxation rates γ_α, we find $F_+ \simeq 0$ and $F_- \simeq 2$. The width Γ_- is found to be real for all values of the Rabi frequency μ_1 (μ_2 treated to first order), which means that no ac Stark shifts occur in this system. Also Γ_- is found theoretically to be rather insensitive to μ_1.

Brewer and Hahn[8] have solved the equations of motion for this resonant three-level case, assuming equal decay rates for the diagonal elements of the density matrix. They find a two-photon resonance, both broadened and shifted, but with the shift vanishing for equal Rabi frequencies $\mu_1 = \mu_2$ and small symmetric detuning.

These predictions have been investigated in two ways. First, we have recorded the two-photon signal as a function of μ_1 and μ_2. Even for Rabi frequencies $\mu_i \simeq 5\gamma_\alpha$, $\mu_1 = \mu_2$, we do not find any shift of the two-photon resonance within 1×10^{-8}, independent of detuning within the velocity range shown in Fig. 3. This fact is supported by the experimental verification of $F_+ = 0$, which means that coherent processes are solely responsible for the population monitored in the $4d'$ level. However, this, in turn, means that no population builds up in $3p'$, which is exactly the condition for a vanishing ac Stark shift, the reason being that $\gamma_{3p'} > \gamma_{4d'}$. These points can be studied further by observing the population in the $3p'$ level. The population is proportional to[12] two terms, (i) a direct Doppler-broadened absorption, $3s$-$3p'$, and (ii) a coherent Doppler-free contribution.

In Fig. 4 are shown recordings indicating the sharp, two-photon, Doppler-free resonance sitting on a Doppler-broadened background absorption. As we did not find any shifts of the two-photon resonance, the shift of the $3p'$ level can be deduced by measuring the shift of the Doppler feature relative to the two-photon signal as a function of beam energy. In the power part of Fig. 4, this detuning versus beam energy is shown. No deviation from a pure Doppler shift is observed, once again showing the stability of this system against ac Stark shifts.

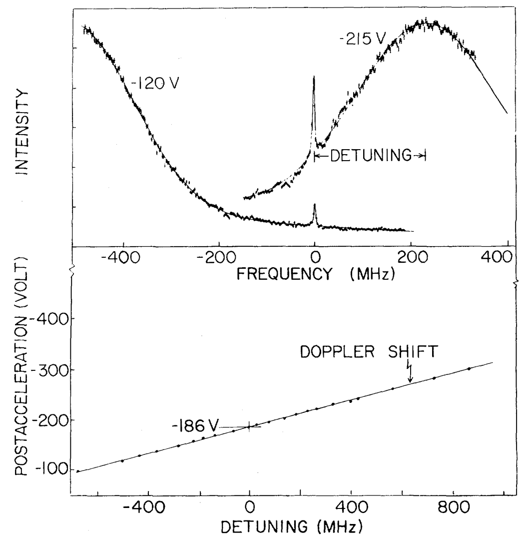

FIG. 4. Experimentally obtained signal seen in the decay $3p'$-$3s$ at 6677 Å (upper part). Within our experimental accuracy, the energy separation between the two-photon and the Doppler-broadened resonance ("detuning") arises solely from the Doppler shift, indicating no ac Stark shift of the $3p'$ level (lower part).

Besides the modest broadening effects observed, the experimental linewidth is composed by (i) the finite laser bandwidth of 1 MHz, (ii) the residual second-order Doppler broadening of ~1 MHz, (iii) the transit-limited width $\gamma_{3s} \simeq 0.3$ MHz, and finally (iv) the excited-state lifetime $\gamma_{4d'} = 3$ MHz with the residual Earth magnetic field eliminated by external coils.

What are the ultimate resolution limits in two-photon absorption in fast beams? Two factors interact, i.e., the transit-time limit and the second-order Doppler broadening, the first favoring low-velocity dispersion. With some care, the second-order Doppler broadening can be reduced to 10 kHz and the transit-time limit to 100 kHz. Many harmonic three-level atoms or ions can be "created" by use of this technique, and its inherent high resolution makes it desirable from both an applied spectroscopic and a metrologic point of view. By adding a second stabilized dye laser to the experiment, tuned to the red-shifted $3s$-$3p'$ transition, the second-order Doppler shift can be isolated uniquely to within a few parts in 10^{-6}. This would constitute a major improvement over previous measurements of this kind.

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Self-Organization in Korteweg-de Vries Turbulence

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Turbulence describable by the Korteweg–de Vries equation with a frictional dissipation is shown to self-organize to a soliton.

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Recently self-organization of turbulence (or random state) is attracting interest in many areas. For example, the three-dimensional, low-β magnetohydrodynamics fluid with incompressible perturbation is found[1,2] to self-organize to a state in which the magnetic flux density B satisfies the field equation $\nabla\times B=\lambda B$. This equation is derived by applying the variational principal to minimize energy, $E=\int B^2 dV$, with the constraint of a constant magnetic helicity $H=\int A\cdot B dV$. Here $B=\nabla\times A$ and λ is the Lagrange multiplier. The philosophy behind this conjecture is that in the case of a large Reynolds number, the system evolves to a quasistationary state through nonlinear mode couplings that minimize E with H kept constant through a selective dissipation of E.

A similar argument was successfully applied by Bretherton and Haidvogel[3] as well as by Matthaeus and Montgomery[4] to the self-organization of a two-dimensional incompressible fluid. They have demonstrated using computer simulations that the system evolved to a state in which the enstrophy $U=\int(\nabla\times v)^2 dV$ was minimized with the constraint of a constant energy $W=\int v^2 dV$.

The self-organization can loosely be defined as a process of formation of ordered structure from a disordered structure in a nonlinear and dissipative system. By taking the common features of the above two examples, here we define self-organization as a process in which (1) a selective dissipation exists among conserved quantities such that a higher-order conserved quantity dissipates faster, and (2) the energy spectrum transfers to smaller wave numbers.[5] Although the conditions (1) and (2) are not necessarily independent, it is convenient to use the both criteria since condition (2) warrants formation of a coherent structure while condition (1) may be used to construct an appropriate field equation that represents the self-organized state. In this Letter we show that the Korteweg–de Vries (KdV) equation with a frictional dissipation satisfies these two criteria, and hence it can form a self-organized state. The self-organized state appears in the form of a soli-

