Direct observation of Rabi oscillations in transmission signal of atomic vapor under continuous-wave laser excitation

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We have studied the temporal behavior of the atomic absorption signal under resonant excitation with a continuous-wave laser radiation. Measurements done for D$_2$ line of $^{85}$Rb with $\approx$ 1 ns temporal resolution have shown irregular oscillatory behavior of the transmission signal, which becomes well pronounced for high laser power, and disappears when the laser is tuned off-resonance. Application of the fast Fourier transform analysis of the transmission signal reveals power-dependent frequency peaks, which are shown to be associated with Rabi frequency.

Resonant interaction of narrow-linewidth cw laser radiation with atomic vapor (notably, alkali metal vapor) is intensely studied in the past decades, driven by fundamental interest and important emerging applications. Most of these studies deal with a steady-state regime of interaction of atomic ensemble with resonant light required for establishment of the relevant processes. The establishment of a steady-state regime implies the onset of a dynamic balance between elementary processes (e.g. absorption and emission), which leads to the invariance of the average level of an atomic signal in time under invariable excitation conditions. But a question remains: is the stationary signal time-independent in a short time scale? The dynamic contribution of elementary processes (Rabi cycle) can be revealed in measurements with high temporal resolution.

Rabi oscillations were directly observed in microwave domain in many experiments, meanwhile this task becomes much more problematic in the optical frequency range, notably for the transitions with large dipole moment. Optical Rabi oscillations with a half period as short as 1 ns have been detected in single nitrogen vacancy centers in diamond [1]. Direct detection of the time-resolved Rabi oscillations in the coherent transient response of the resonance fluorescence of a single charged InAs quantum dot has been reported in [2]. Ultrafast Rabi oscillation of an atom ensemble in Gaussian spatial distribution has been investigated in [3] using the cold atomic rubidium vapor spatially confined in a magneto-optical trap. All these experiments were done with pulsed laser excitation.

In this Letter we report on the temporal structure in atomic absorption signal under continuous-wave laser excitation observed with fast detection and processing technique. The experiment is done for D$_2$ line of rubidium in a simple configuration with atomic vapor cell.

The sketch of experimental setup is shown in the upper panel of Fig. 1. It comprised of a simple main arrangement for dynamic recording of the radiation transmitted through a rubidium vapor cell, and a more complex auxiliary arrangement for tuneable locking of laser radiation frequency.

An unfocused $\approx$ 2-mm-diameter linearly-polarized beam from a single-frequency free-running cw diode laser (wavelength 780 nm, light power 30 mW, linewidth 15 MHz) was directed into a 70 mm-long, 10 mm-diameter sealed cylindrical cell with a side arm containing natural rubidium. An optical isolator was used to prevent laser cavity effect imposed by reflections; the power of laser radiation entering the cell was controlled by a circular variable neutral density filter. The vapor-cell temperature was kept at 27.8°C, which corresponds to a number density of Rb atoms $N = 9.27 \times 10^{10}$ cm$^{-3}$. The radiation transmitted through the cell was detected by a Newport 818-BB-21A optical biased receiver (PD1) with 1.2 GHz bandwidth and 0.5 ns rise/fall time. This photodetector with a sensor diameter of 0.4 mm was mounted on a translation stage enabling adjustment of its position across the transmitted beam aperture.

Signal from the photodetector PD1 was monitored by the Tektronix TDS3032B digital storage oscilloscope...
with 300 MHz bandwidth and 2.5 GS/s sample rate per channel. An integrated fast Fourier transform (FFT) module TDS3FFT built in the oscilloscope allowed to perform a real-time Fourier analysis of the photodetector signal.

Small fraction of the laser beam was branched onto a dichroic atomic vapor laser lock (DAVLL) scheme [4] allowing realization of locking the laser radiation frequency to atomic resonance line employing linear magnetooptics. The laser frequency was locked to the chosen atomic resonance via a feedback loop controlling the injection current of laser diode, with a dispersion-shaped error signal formed by the differential amplifier.

The following procedure was employed for the measurements. First, the laser was locked to the Doppler-overlapped atomic transition group $^{85}$Rb $F_y=3 \rightarrow F_e=2,3,4$ of the Rb D$_2$ line (see lower panel of Fig. 1), and absorption (PD1) signal was recorded for a 2 µs interval (no averaging, 10000 measurement points with the step of 0.2 ns). The measurement data were stored together with the real-time Fourier transform spectrum generated by an FFT module operating in linear root-mean-square output and rectangular window mode, which assures maximum frequency accuracy. Afterwards, the same measurements were done with the laser radiation frequency tuned to the off-resonance position.

The measurements were done for 8 values of the laser radiation power $P_L$: 2, 5, 8, 11, 14, 17, 20, and 23 mW. We should note that the real power entering the cell was 1.18 times less because of reflection from the sapphire window cut across the c-axis to avoid birefringence. The net absorption of the transmitted beam at atomic resonance ranged from 30.5% for the lowest power to 9.55% for the highest power. Throughout the measurements, the PD1 photodetector sensor was carefully aligned to the center of the transmitted beam.

The measurements results for $P_L = 2$ and 23 mW are presented in Fig. 2. One can clearly see an irregular oscillatory behavior of the transmission signal when the laser radiation frequency is tuned to the atomic resonance (graphs a and e), with the magnitude increasing with $P_L$. Noteworthy, the temporal structure gets out-vanished already at integration time of 100 ns (see red lines in graphs a and e). The oscillations practically disappear when the laser frequency is driven out of the resonance (graphs c and g). Application of a fast Fourier transform (right column graphs) allows revealing the frequency spectrum of the signal, which exhibits a peak behavior, with a maximum frequency rising with $P_L$.

Experimental dependence of the peak frequency of the FFT signal on the incident laser radiation power is plotted in Fig. 3. Solid dots present the averaged values of 5 measurements for each value of $P_L$, with an error of ±7.5%. The square-root-shaped course of this dependence allows us to suppose that the oscillatory behavior of the transmission signal should be attributed to dynamic Rabi cycle. To verify this supposition, we have made estimates for the expected generalized (effective) Rabi frequency $\tilde{\Omega}$:

$$\tilde{\Omega}_{ij} = \sqrt{\frac{d_{ij}^2 P_L}{\epsilon_0 n c \hbar^2 S}} + \Delta^2. \quad (1)$$

(see e.g. [5]), where $d_{ij}$ is the transition dipole moment, $\epsilon_0$ is the vacuum permittivity, $n$ is the refractive index, $c$ is the speed of light, $S$ is the effective area of the laser beam cross-section, and $\Delta$ is the detuning.
of the laser radiation frequency from the atomic transition (including also Doppler broadening and the spectral linewidth of laser radiation when it exceeds transition natural linewidth).

The main contribution in the Doppler-overlapped ($\approx 640$ MHz FWHM) hyperfine transition group $^{85}$Rb $F_g=3 \rightarrow F_e=2,3,4$ gives the cycling transition $F_g=3 \rightarrow F_e=4$, having a strength factor, which exceeds the strength factors of $F_g=3 \rightarrow F_e=3$ and $F_g=3 \rightarrow F_e=2$ by factors of 2.3 and 8.1, respectively. As a result, also the frequency of this transition is the closest to the transmission minimum ($\approx 40$ MHz). For this reason, in calculations we have used for $d_{ij}$ the effective dipole moment $1.659 \times 10^{29}$ C/m of the $F_g=3 \rightarrow F_e=4$ transition [6]. The calculated dependence is plotted in Fig. 3 by a solid line. The best fit has been obtained with the effective detuning $\Delta = 85.4$ MHz, which is realistic given the experimental uncertainties (contributions from other transitions, laser beam profile, thermal motion of atoms across the beam, Doppler broadening, laser linewidth, relaxations, etc.).

Moreover, it is important to note that the frequency corresponding to the maximum of FFT signal does not correspond to the maximum of Rabi frequency. Analyzing the shape of FFT spectrum, one can say that the most intense interaction occurs with the velocity group of atoms, which are in exact resonance with the laser field, and these atoms contribute to the far high-frequency wing of the FFT profile. The most intense FFT signal corresponds to the contribution of atoms with the mean longitudinal thermal velocity. One may say that the FFT profile somehow reflects Maxwellian velocity distribution. The shape and width of the FFT spectrum profile can be also affected by the spatial distribution of intensity across the photodetector sensor area, the optical length of the cell, the vapor density, the presence of buffer gas or antirelaxation coating.

Finally, let us comment on the ”permanence” of the observed Rabi oscillations for continuous-wave excitation of atomic vapor. Here we should note that each particular atom interacts with the laser field within a limited time (a $\mu$s-range time of flight of an atom through the laser beam). As a result, at each moment of time the overall dynamic equilibrium of atoms in the laser beam is maintained by a rapid succession of departing and arriving atoms, resembling the pulsed interaction regime.

In conclusion, we have observed and qualitatively analyzed appearance of a temporal structure in atomic vapor transmission signal. This observation became possible thanks to usage of an ultrafast detection system with small-aperture photodetector, and application of FFT signal processing. As a follow-up action, we plan to perform a comprehensive and detailed experimental study combined with theoretical modeling, which will properly account for all the physical processes involved.

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