New Type of High-Resolution Spectroscopy with a Diode Laser

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(Received 17 December 1990)

A phenomenon observed in a simple experiment with a diode laser is found to be a basis for a powerful spectroscopy. In this spectroscopy, the laser frequency is neither scanned nor modulated. High-resolution spectra in a wide radio-frequency range, such as Zeeman and hyperfine spectra in both of the ground and the excited states, are observed simultaneously by frequency analyzing the intensity fluctuation of the light transmitted through a sample cell. Demonstrating experiments are carried out with respect to the D_1 and D_2 lines of Cs and Rb atoms.

PACS numbers: 32.80.Bx, 32.30.−r

In this paper, we report on a new type of high-resolution spectroscopy with a diode laser, which is considerably different from other spectroscopic methods developed so far. Above all, the optical system is extremely simple; it consists simply of a diode laser, a sample cell, a fast photodetector, and a radio-frequency spectrum analyzer. In this spectroscopy, it is unnecessary to scan or modulate the laser frequency externally in order to get atomic spectra. It is enough to tune the laser frequency roughly to the Doppler-broadened absorption line. Atomic spectra in the frequency range from tens of kHz to several GHz, such as Zeeman and hyperfine spectra in both of the ground and the excited states, can be obtained by frequency analyzing the intensity fluctuation of the light transmitted through the atoms.

This spectroscopy is based on the characteristics of a diode laser. First, the output amplitude of a diode laser is generally very stable, compared with other lasers. As recently shown by Machida, Yamamoto, and Itaya [1], the amplitude fluctuation becomes less than the shot-noise limit, i.e., the output is amplitude squeezed, when it is driven by a constant-current source. On the contrary, the frequency fluctuates at random, i.e., with a very short correlation time, which results in a broad spectral line, the half-width being typically tens of MHz but the wing extending up to several GHz from the line center.

In the course of simple absorption spectroscopy of alkali vapor with a diode laser, we found that the stable intensity of the diode laser beam became quite noisy when it was transmitted through the vapor. Such excess noise has also been observed by Haslwanter *et al.* [2] and Ritsch, Zoller, and Cooper [3]. The noise amplitude becomes maximum when the laser frequency is resonant with the atomic absorption lines. Figure 1 shows the intensity change of the light transmitted through Cs atoms, observed when the laser frequency was scanned slowly through the absorption line from the $F=4$ level in the ground state to the $6P_{1/2}$ state. One can see that the noise amplitude is very large at resonance, larger than the change of the average light intensity. Since the output of the diode laser was still very stable even in such a case, we found that the intensity noise was not due to the instability of the diode laser by the feedback of the laser light or spontaneous emission from atoms. We found in this way that the intensity fluctuation in the transmitted light was a response of the atoms to the laser field with the frequency fluctuating at random, i.e., with a wide frequency spectrum. Therefore, we considered that the intensity noise must contain such information about the interacting atoms as energy splittings and lifetimes of associated states.

Figure 2(a) shows the experimental setup to observe Zeeman resonance signals. The diode laser used here was an ordinary Fabry-Pérot type with output power of 3 mW, operating on a single mode at the Cs D_1 (894 nm), Cs D_2 (852 nm), Rb D_1 (794 nm), or Rb D_2 (780 nm) lines. The laser temperature was controlled within 1×10^{-4}°C by a servo system with a Peltier element, and the driving current was also controlled down to less than 1 μA. Thus, we could obtain a long-term fluctuation of a few MHz [4]. The output of the diode laser was applied to the cell containing Cs or Rb vapor, which was placed in a transparent oven in order to maintain the cell temperature at 30–60°C. As in the transverse optical pumping experiment [5], a static magnetic field directed per-

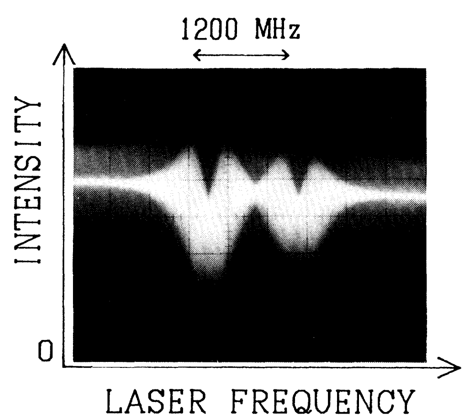

FIG. 1. Intensity change of the diode laser beam transmitted through Cs vapor observed when the laser frequency was slowly scanned through the D_1 line.

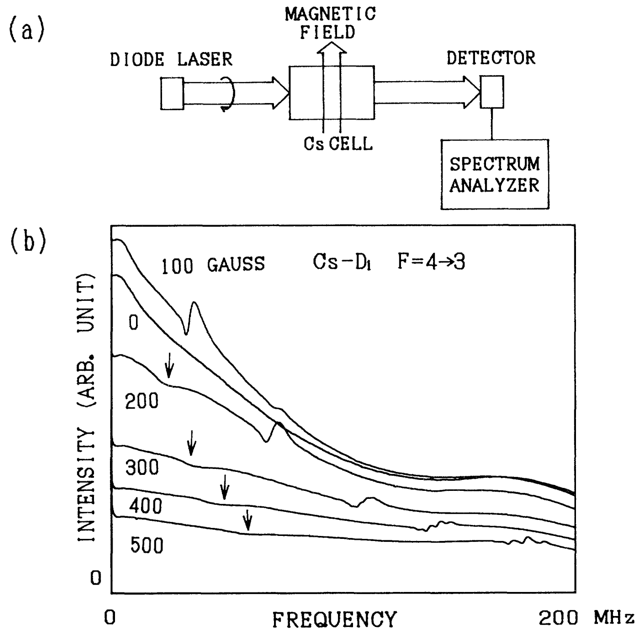

FIG. 2. (a) Experimental setup to observe Zeeman resonances, and (b) the obtained frequency spectrum of the fluctuating transmitted light intensity, for various values of magnetic field. The weak resonance indicated by an arrow is due to Zeeman coherence in the excited state.

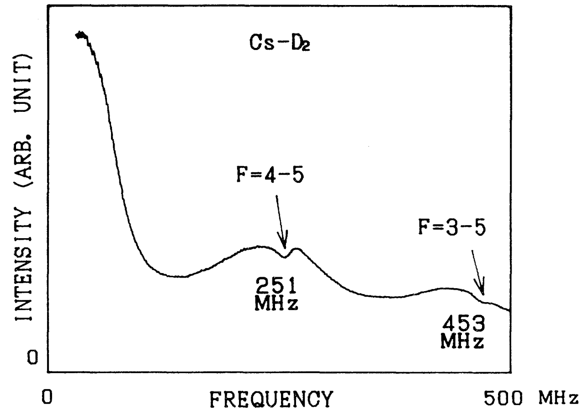

FIG. 3. Observed hyperfine resonances for $F=4\rightarrow5$ and $F=3\rightarrow5$ in the $6P_{3/2}$ state in the frequency spectrum of the transmitted light intensity.

pendicularly to the laser beam was applied to the cell by a set of Helmholtz coils. The incident light was circularly polarized, and light transmitted through the cell was detected by an avalanche photodiode, which had frequency response up to 1.3 GHz. The output of the detector was directed to the frequency analyzer to obtain the power spectrum of the fluctuation of the transmitted light intensity.

Figure 2(b) represents the recorder traces showing the intensity noise spectrum of the light transmitted through Cs vapor. In this case, the laser frequency ω was tuned to about the center frequency ω_0 of the Doppler-broadened transition of Cs from the $F=4$ level in the ground state to the $F=3$ level in the $6P_{1/2}$ state. In Fig. 2(b), we see the broad background noise spectrum. This noise spectrum was found to extend over the limit of the frequency response of the photodiode used, 1.3 GHz, and the frequency at the peak was given by the detuning of the laser frequency from the line center. When the laser frequency was completely off resonant, the noise level was reduced to less than $\frac{1}{1000}$ of above spectrum. On the slope of the background spectrum, we see clearly a sharp spectrum whose frequency is given by the Larmor frequency in the ground state of Cs, i.e., 366 kHz/G. When the magnetic field is stronger than 300 G, this spectrum is decomposed into several lines, because of larger inequality of the Zeeman splittings. The resonance width is given approximately by the inverse of the transit time of atoms across the laser beam. We can also see another weak and broad resonance at one-third of the frequency of the strong resonance. Since the g factor in the $6P_{1/2}$ state is $\frac{1}{3}$ of that in the ground state, and the resonance width is given approximately by the natural width, this weak resonance is apparently due to Zeeman coherence in the excited state.

The resonances at the hyperfine splittings in the excited states could also be observed in the background noise spectrum, in the same system as in Fig. 2(b). In this case, the magnetic field was removed, and the linearly polarized output of the diode laser was directly applied to the atoms. Figure 3 shows the observed spectrum for the case that the laser frequency is tuned to the D_2 line of the $6P_{3/2}$ state of Cs, where we can see sharp spectra at the frequencies corresponding to the hyperfine splittings in the $6P_{3/2}$ state. The width of these spectra is given by the natural width. In this way, we could observe all the hyperfine spectra of the $P_{1/2}$ and $P_{3/2}$ states of the first excited states of ^{133}Cs, ^{85}Rb, and ^{87}Rb. The obtained hyperfine constants agreed with the previous values [6] measured by using the techniques of atomic beam or level crossing, within the error of less than 1 MHz.

In Fig. 3, we also see that the background spectrum has broad structures. We found that these structures were the effects of the holes burned in the velocity distribution of the atoms. To show this more clearly, we made an experiment with Cs atoms, to which two coaxial laser beams from different lasers were applied simultaneously. One laser beam was used as a probe beam which was tuned on the wing of the absorption line from the $F=4$ level in the ground state to the $F=3$ level in the $P_{1/2}$ state, and the intensity fluctuation of the transmitted beam was frequency analyzed. The other beam was used as a pump beam, which was tuned so as to excite the atoms in the $F=4$ level in the ground state to the $F=5$, 4, and 3 levels in the $P_{3/2}$ state. Figure 4(a) shows the

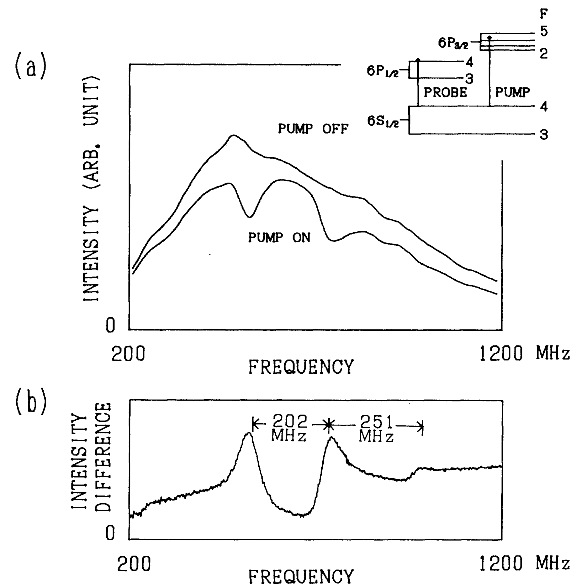

FIG. 4. (a) Frequency spectra of the transmitted probe beam intensity, in the absence and the presence of the pump beam. The difference of these spectra is shown in (b). In this experiment, the frequencies of the probe and pump beam were fixed to the D_1 and D_2 lines of the Cs atom, respectively.

spectra obtained in the absence and presence of the pump beam, and 4(b) shows the difference of these spectra. We can see the hole-burning spectra corresponding to three transitions induced by the pump beam. The resonance shapes are not simple, the widths of which are roughly given by the sum of spectral widths of the two diode lasers, about 30 and 40 MHz in the present case. Similar Doppler-free spectra can also be observed in saturation absorption spectroscopy [7], but it should be emphasized that, in the present case, Doppler-free spectra can be obtained without scanning the frequency of either the probe or the saturating beam through resonances.

The phenomena described above are due to the linear and nonlinear interactions of atoms with a laser field with a stable amplitude and a frequency fluctuating at random. In fact, we found that the intensities of the background noise spectrum had a linear dependence on the incident light intensity I_0 and those of the sharp resonances had approximately a I_0^2 dependence. Recently we studied theoretically with simplified models the situation where a two- or three-level atomic system interacts with a laser field whose frequency fluctuation has an extremely short correlation time, i.e., it is modulated by correlationless spontaneous emission noise and carrier noise [8]. The results could explain the appearance of the hole-burning and Zeeman (hyperfine) resonances. We briefly mention here the outline of the theory (details will be reported elsewhere). When we denote the laser field by $E_L(t)$ and the field from the induced dipole moment by $E_d(t)$, the transmitted light intensity can be expressed by the

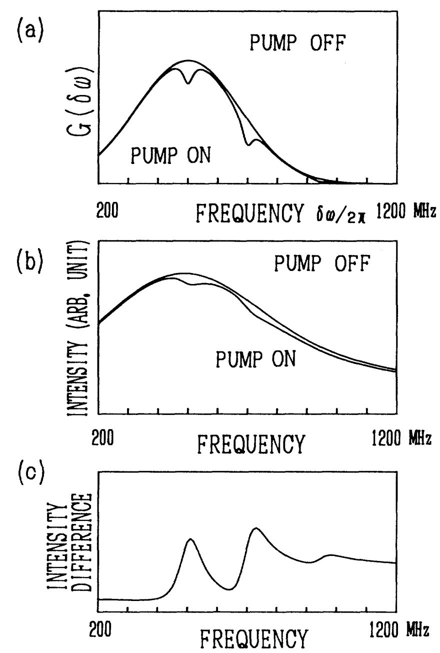

FIG. 5. Theoretical frequency spectra of the transmitted light intensity, corresponding to the experiment with two laser beams (see Fig. 4). (a) The Doppler profile with three holes used in the calculation, where $\delta\omega$ is atomic detuning from the center of the probe laser line. The spectra shown in (b) and (c) correspond to Figs. 4(a) and 4(b), respectively.

square of the sum of these fields, i.e., $|E_L(t)|^2 + 2\mathrm{Re}[E_L(t)^* E_d(t)] + |E_d(t)|^2$. When the atomic vapor is optically thin, the term $2\mathrm{Re}[E_L(t)^* E_d(t)]$ is the main source term of the generation of the intensity fluctuation. The dipole field from the group of atoms with a particular Doppler-shifted resonance frequency ω_a has a narrow natural width. So, when the laser frequency is detuned from the center of the absorption line, the ensemble of atoms is excited through the wing of the laser spectrum. In such a case, $E_d(t)$ will have a spectrum approximately given by the Doppler distribution $G(\omega_a)$. In this way one can see that the background noise produced by the heterodyne beat of $E_L(t)$ and $E_d(t)$ will have a similar spectral shape as G. When the distribution function $D(\omega_a)$ has three holes as shown in Fig. 5(a), which is the case of our experiment with two laser beams, the theoretical results shown in Figs. 5(b) and 5(c) agree well with the spectra shown in Figs. 4(a) and 4(b).

As an example of sharp resonances, let us consider here the Zeeman resonance in the ground state. The atoms with a particular velocity see the pulsed circularly polarized light, and transverse magnetization, i.e., Zeeman coherence, is produced by optical pumping. The amplitude and phase of this precessing magnetization depends on the velocity of the atoms. However, the sum of the Larmor frequency components in $E_L(t)^* E_d(t)$ over velocity does not cancel, because the detection of Larmor precession is also velocity dependent. The precession signal is mainly produced by the off-resonant atoms, i.e., the atoms interacting with the wing of the laser line, through dispersion. This makes sense with the experimental fact that the width of the Zeeman resonance does not have a significant intensity dependence. So, the mechanism is basically similar to that for the techniques of Stark switching and frequency switching developed by Brewer and co-workers [9,10]. We made a numerical calculation with a three-level system, two levels being the ground-state Zeeman sublevels, and obtained results explaining the experimentally observed resonances, even their line shape.

We have made more active experiments using the above-mentioned phenomenon. The first system studied this time is one modified from that in Fig. 2, where the detected signal is fed back to the driving current of the diode laser, i.e., to the laser frequency. In a second system, the transmitted light is fed back directly to the diode laser. We found that, in both systems, a continuous oscillation occurs at the Larmor frequency of the ground state, when the feedback gain exceeds some critical values. This oscillation appears as the frequency modulation of the diode laser. The frequency-modulated light sustains the spin precession of the atoms in these systems, as seen in the spin-related optically bistable [11] and tristable [12] systems. The details of these experiments are also to be reported in a separate paper.

In this paper, we have reported a phenomenon observed in a simple experiment with a diode laser. We found that this phenomenon could be used as a high-resolution spectroscopy, which provides at one time much precise information about the atomic states covered by the laser spectral line. This spectroscopy may become very useful for the species and states whose precise spectra are not known in advance. Finally, it should be mentioned that, if we detect a particular frequency component of the transmitted light intensity and scan the laser frequency through the absorption line, we can get the Doppler-broadened spectrum with the same shape as the one observed in the frequency-modulated spectroscopy [13].

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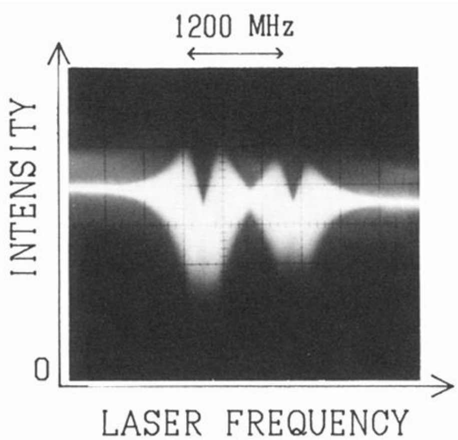

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