Radiative transfer of a nonequilibrium atomic velocity distribution between degenerate sublevels of an atomic ground state during optical pumping in an intense monochromatic field

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(Submitted 16 August 1993)
Pis'ma Zh. Eksp. Teor. Fiz. **58**, No. 7, 501–504 (10 October 1993)

Optical pumping on the $^3P_2 \rightarrow {}^3D_2$ ($1s_5 \rightarrow 2p_8$) transition of Ne^{20} in intense monochromatic laser light has been studied experimentally. A coherent radiative transfer of a nonequilibrium atomic velocity distribution between degenerate sublevels of the atomic ground state has been detected. The transfer is to a sublevel which is not interacting with the intense field. The population distribution of this sublevel acquires a sharp peak at the field frequency. The height of this population peak is more than twice the height of the Doppler pedestal. The coherence of the population transfer makes it possible to directly observe a Stark splitting of sublevels of the excited state in the field of counterpropagating waves. The observed effect can be interpreted as an analog of coherent trapping of population in a nonabsorbing atomic state in an optical-pumping cycle, with the possibility of an arbitrary selection of the velocity of the atom. The results open the way to an effective method for selecting polarized atoms and for controlling their motion.

1. The nonequilibrium atomic velocity distribution which arises in the interaction of an intense optical field with an ensemble of atoms is being studied by laser saturation spectroscopy.[1] An analysis of saturation processes carried out for the cases of simple and degenerate two- and three-level atomic systems reveals that some characteristic features (resonances) appear in the absorption and emission spectra for various combinations of a strong (saturating) field and a weak (probe) field in terms of their relative propagation directions and also their frequencies and polarizations.

Another possibility for sub-Doppler spectroscopy is the method of velocity-selective optical pumping.[2,3] A distinguishing feature of the atomic systems which have been studied by the optical pumping method is that the relaxation processes in the system of degenerate sublevels of the ground state are slow. Another distinguishing feature is the low pump intensity sufficient to saturate the transition. As a result, there is a redistribution of population among degenerate sublevels of the atomic ground state. In several cases there is also a bleaching of the atomic transition to produce light of a given polarization.

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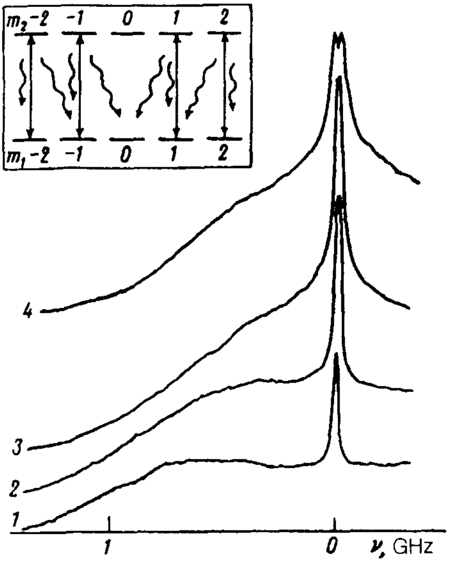

FIG. 1. Shape of the absorption line of a circularly polarized weak wave in the field of a linearly polarized, counterpropagating strong wave for the optically orientable transition ${}^3P_2 \rightarrow {}^3D_2$ ($1s_5 \rightarrow 2p_8$) of Ne^{20} at various intensities of the strong field. 1—3×10^{-2} W/cm^2; 2—6×10^{-2}; 3—1.2×10^{-1}; 4—0.5 W/cm^2. Shown at the upper left is the scheme of stimulated transitions (straight arrows) and spontaneous transitions (wavy arrows) to the ground state (${}^3P_2$) of the neon atom.

The primary relaxation mechanism in optically oriented atomic systems is radiative relaxation from the excited state to the ground state. Accordingly, the theory of optical pumping for light intensities at the laser level requires a self-consistent account of induced and spontaneous emission. As a result, new features of the distribution of population with respect to degenerate Zeeman sublevels of the atomic ground state are identified.[4]

In the present letter we are reporting the first, to the best of our knowledge, experimental results on the observation of a coherent radiative transfer of a nonequilibrium atomic velocity distribution among degenerate sublevels of the ground state under conditions of optical orientation by intense laser light.

2. The experiment was carried out on the ${}^3P_2 \rightarrow {}^3D_2$ ($1s_5 \rightarrow 2p_8$) transition of Ne^{20} ($\lambda = 633.4$ nm), whose ground state is metastable. This transition is an example of a system which can be optically oriented and in which linearly polarized pump light causes the atoms to become redistributed to the $m_1 = 0$ Zeeman sublevel of the ground state as a result of radiative transitions. This effect leads to a bleaching of the transition even at a moderate pump intensity. Figure 1 shows the scheme of transitions. We regarded the primary purpose of this experiment as a task to detect a spectral dependence of the shape of the absorption line for a weak, circularly polarized wave as the result of the transitions $m_1 = 0 \rightarrow m_2 = \pm 1$ in the field of a counterpropagating, linearly polarized, strong wave for the case of equal wave frequencies ("strong" in the sense that the transition is bleached).

The experimental apparatus was of the standard type for experiments on the observation of a saturated-absorption resonance in the field of counterpropagating waves in a gas.[1] The beam from a cw tunable laser using the dye DSM entered a discharge cell with pure Ne^{20}. The neon pressure was 2×10^{-2} torr, the discharge current was held at ~ 15 mA, and the length of the absorbing layer was 5 cm. Under these conditions the density of Ne atoms in the $1s_5$ metastable state was[5] $\sim 5\times 10^9$ cm^{-3}, and the unsaturated absorption coefficient was $\sim 2\times 10-2$ cm^{-1}. The power of the pump light, which was linearly polarized, could be varied continuously from 0 to 30 mW; the corresponding power densities are 0–0.5 W/cm^2. The width of the emission line was $\leqslant 3$ MHz. The counterpropagating, circularly polarized weak field had a constant power $\simeq 0.1$ mW and was directed at an angle $\sim 5\times 10^{-3}$ rad with respect to the strong field. The shape of the absorption line was determined from the changes in the intensity of the weak field as the laser frequency was tuned over the neon absorption line.

3. Figure 1 shows experimental recordings of the absorption line for the weak, circularly polarized wave in the field of the counterpropagating, linearly polarized, strong wave for the optically orientable transition $^3P_2 \rightarrow {}^3D_2$ of Ne^{20}, for various intensities of the strong field. A zero absorption coefficient for the weak field corresponds to the base of the absorption line, as we verified by disconnecting the discharge cell and by tuning the laser frequency.

The intensity of the strong field, which leads to a redistribution of atoms among degenerate sublevels of the ground state in the optical-pumping cycle, is determined by the total bleaching time, which is $\tau \sim 1/\gamma G$ for weak fields and $\tau \sim 1/\gamma$ for strong fields,[2,4] and also by the relation between τ^{-1} and the rate of depolarizing collisions. Here γ is the relaxation rate of the upper level, and G is the transition saturation parameter. For our experimental conditions, a bleaching corresponding to the emptying of the Zeeman sublevels with $|m_1| = 1, 2$ is reached at a pump power density $I_{p,\min} \geqslant 2\times 10^{-2}$ W/cm^2. This figure was found from the behavior of the absorption coefficient for the strong field as a function of the intensity of the latter. The shape of the absorption line for the weak field was studied at $I_p > I_{p,\min}$.

The absorption line observed for the weak field is a narrow resonance against the background of a broad Doppler pedestal, formed by absorption directly from the $m_1 = 0$ magnetic sublevel. The reason for the resonant increase in the absorption at the center of the line is a radiative transfer of population from the $|m_1| = 1, 2$ magnetic sublevels to the $m_1 = 0$ sublevel in the optical-pumping cycle in the intense electromagnetic field. An increase in the intensity of the strong field leads not only to an increase in the height of the resonance, i.e., an increase in the population of the $m_1 = 0$ sublevel, but also to a field-induced broadening of this resonance. The ratio of the height of the resonance to the height of the Doppler pedestal reaches 2.5 at a pump power density $I_p \simeq 3\times 10^{-2}$ W/cm^2. The width at half-maximum of the resonance, Γ_p, increases from 20 MHz at $I_p \simeq 3\times 10^2$ W/cm^2 to 45 MHz at $I_p \simeq 1.2\times 10^{-1}$ W/cm^2. The field-induced broadening of the resonance is a nonlinear function of the pump intensity. Since the strong and weak fields are at the same frequency, we can assert that there is a coherence in the absorption of the strong and weak fields during the radiative transfer of population between Zeeman sublevels of the ground state of this optically orientable atomic system in the absence of depolarizing collisions.

A further increase in the intensity of the strong field leads to a Stark splitting of the observed resonance of the population of the $m_1 = 0$ magnetic sublevel into two components. This splitting is convincing evidence that the transfer of nonequilibrium population to a sublevel which does not interact with the field is a coherent process. The magnitude of the Stark splitting is $\simeq 25$ MHz at $I_p \simeq 0.5$ W/cm^2 (Fig. 1). The ratio of the height of the resonance to that of the Doppler pedestal typically begins to fall off at $I_p \simeq 1.5 \times 10^{-1}$ W/cm^2, reaching a constant value (~ 2) at $I_p \gtrsim 0.2$ W/cm^2.

Interestingly, an example of an optically orientable scheme of transitions was analyzed in Refs. 1 and 6 (orientation was ignored). It was shown there that the absorption coefficient κ for a weak wave at a frequency ω_2 in the presence of a counterpropagating strong wave at ω_1, with the opposite polarization, under the condition $\omega_1 = \omega_2 = \omega$, is

$$\frac{\kappa}{\kappa_0} = \exp\left[-\left(\frac{\Omega}{kv}\right)^2\right]\left\{1 - G\frac{\gamma_1 \Gamma}{(2\Gamma)^2 + \Omega^2}\right\},$$

where $\Omega = \omega_{21} - \omega$, ω_{21} is the transition frequency, γ_1 is the relaxation rate of the ground level, 2Γ is the homogeneous width of the transition, G is the transition saturation parameter, and κ_0 is the unsaturated absorption coefficient.

It can be seen from the behavior found that there is an ordinary Doppler lineshape at $\Omega \gg 2\Gamma$, governed by the population of the Zeeman level which is not affected by the strong field, while at $\Omega < 2\Gamma$ there is a dip in κ due to the peak in the population in the common upper level. This result is diametrically opposed to the result which emerges from our experiments, and it indicates that optical pumping is important to an analysis of real atomic systems.

4. In summary, this study has shown that optical orientation of atoms in intense laser fields results in a coherent radiative transfer of a nonequilibrium atomic velocity distribution to one of the degenerate sublevels of the atomic ground state. A very important point is that this sublevel is one which is not subject to the effect of the strong field. In this regard, the results can be interpreted as an analog of coherent population trapping in a nonabsorbing state.[7] A distinguishing feature here is that the trapping occurs in an optical-pumping cycle in which there is the possibility of an arbitrary selection of the velocity of the atoms. We regard these results as pertinent to the production of polarized atoms, to the control of their motion, and to their cooling.

We have deliberately avoided taking up several important questions which go beyond the scope of this letter, e.g., the effect of depolarizing collisions on the shape of the absorption line for the weak field, the dynamic Stark effect, and the shape of the Doppler pedestal. We would also like to point out that the lack of a theory for optical pumping for arbitrary field intensities complicates a quantitative interpretation of these results.

We sincerely thank I. M. Beterov, A. M. Tumaĭkin, A. V. Taĭchenachev, and V. I. Yudin for useful discussions, including discussions of these results. This work was supported by the Russian Universities program.

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Translated by D. Parsons