Velocity-selective optical pumping and Doppler-free spectroscopy

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We describe a spectroscopic method combining saturated absorption and Zeeman optical pumping. A weak expanded monochromatic laser beam creates a velocity-correlated orientation in an atomic level without optical saturation. A polarization-modulated counterpropagating detection beam provides signals related to simple atomic observables. Combination with magnetic resonance techniques is described. We have applied this method to metastable states of Ne, with nuclear or electronic orientation. Effects of velocity-changing, depolarization (Kastler- or Dehmelt-type pumping) or metastability exchange collisions are discussed.

Recently, some interest has been given to Doppler-free spectroscopy based on optically induced anisotropy in gaseous samples. Wieman and Hänsch[1] developed a method called "polarization spectroscopy". Another similar technique is "polarization labeling" of molecular spectra.[2] In a recent experiment,[3] the birefringence induced by a linearly polarized intensity-modulated saturating beam was detected by Keller and Delsart. Also, Colomb and Dumont[4] extracted the laser-induced alignment signal from their saturated absorption spectrum by modulating the angle between pump and probe polarization. All these experiments involved saturation of an optical transition, and were therefore of the saturated absorption type.

We report here on a high-resolution laser spectroscopy method without saturation, combining Zeeman optical pumping, velocity selective excitation and detection, and possibly magnetic resonance in an absorbing gas. A circularly polarized laser beam is used to induce anisotropy in the gas, as in ordinary optical pumping. Nevertheless, due to the narrow bandwith of the laser, a strong correlation is introduced between atom velocities and internal variables (orientation, alignment), unlike conventional optical pumping. The method is also distinct from saturated absorption since the atomic observables are created by a pumping beam that is not strong enough to saturate the optical transition ($\vec{D}\cdot\vec{E}\ll\gamma$, where $\vec{D}$ is the atomic transition dipole moment, $\vec{E}$ is the laser electric field, and γ is the spontaneous emission rate). In other words, the induced optical coherence is always completely negligible, and the atomic observable evolve according to rate equations rather than Bloch equations. Therefore, the evolution of these observables can exactly be described by application of the theory of optical pumping[5,6] to the case of velocity selective interaction with the light beam, without resorting to perturbation series. Another consequence of the absence of optical saturation is that this method provides a Doppler-free resonance absorption line essentially without power broadening of the excited state. This is distinct from the saturation spectroscopy techniques quoted in the introduction.

As discussed below, the method can be a useful tool in the study of velocity diffusion of atomic observables (orientation, etc.) in relatively long-lived atomic levels. Its selectivity concerning the choice of these observables, further increased by the combination with magnetic resonance techniques, makes the results easier to interpret theoretically for comparison with collision calculations.

A possible experimental scheme is shown in Fig. 1. A monochromatic laser beam is expanded in order to avoid optical saturation and provide homogeneous pumping over a large volume (~50 cm^3 in our experiment) of the sample cell. A $\lambda/4$ – retarder in the beam gives circular polarization. This results in a velocity-dependent atomic orientation $\langle J_z\rangle(v_z)$, where v_z is the atomic

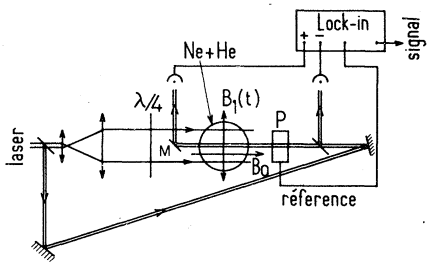

FIG. 1. Experimental arrangement for velocity selective optical pumping. The laser beam is expanded by a telescope and σ^+ -polarized by a $\lambda/4$ plate. The polarization of the probe beam is modulated by the device P (pump beam shadow effects of mirror M are discussed in the text). Differential detection improves the signal-to-noise ratio. A longitudinal magnetic field $\vec{B}_0$ and a transverse rf field $\vec{B}_1$ may be applied for magnetic resonance.

velocity along the laser beam direction.[7] In the absence of atom-atom collisions, a single (Doppler-shifted) velocity group is oriented. For detection, a weak collimated beam is injected as a counterpropagating probe. Geometrically it only interacts with the atoms which are in the very central part of the pump beam, and therefore experience a very-well-defined pumping light intensity. A modulating device P is used to produce a modulated circular polarization $\sigma^{\pm}$ of the probe beam. Thus only the pure orientation $\langle J_z \rangle$ of the probed velocity group will be observed in phase lock-in detection of the probe intensity modulation.[8]

Tuning the probe beam frequency over the Doppler width of the atomic resonance line gives a narrow Lorentzian-shaped signal of natural linewidth at low laser intensities (we assume here that the laser linewidth is small compared to the inverse excited-state lifetime). If both the pump and probe beams come from the same laser and therefore have the same frequency (as in Fig. 1), the narrow resonance appears at the Doppler line center (atoms with zero v_z velocity). In addition, if the atoms undergo collisions which change v_z without entirely destroying their orientation $\langle J_z \rangle$, a diffusion of orientation takes place to other velocity groups within the Doppler width. When the mean-free time τ_c between velocity-changing collisions is much shorter than the orientation relaxation time T_1, the detected spectrum will consist of the superposition of a narrow Lorentzian component and a broader background. This is very similar to what is frequently observed in saturated absorption spectroscopy, although with the present method the extraction of information on collisions from the shape of the curves will be substantially less complicated. This is because saturated absorption spectroscopy necessarily involves population of two atomic levels as well as optical coherences between them. Therefore, any rigourous calculation of the collision effects requires the knowledge of (at least) two atomic potential curves. Also, in saturated absorption measurements, the height and shape of the broad background depend on the value of T_t (interaction time of the atoms with the beam), i.e., on the pumping beam geometry. This is not the case here: T_t is very long, and the background shape depends only on purely collisional time constants.

We tested this method on various isotopes of Ne using several resonance lines linking one of the metastable levels $^3P_2$ or $^3P_0$ of the $2p^5 3s$ configuration to more highly excited states of the $2p^5 3p$ configuration. Our monochromatic light source was a pressure scanned cw dye laser. Single-mode operation was achieved with a "double Michelson" device. This laser has a rather good efficiency ($\simeq 10\%$; single-mode power $\gtrsim 500$ mW), and can easily be tuned over more than 1 cm^{-1}. A preliminary version of it has been described elsewhere.[9] A dye mixture of R 6G and R 101 gave an optimum power near 640 nm, and pure R 6G was used for shorter wavelengths. Nevertheless, since only low pumping power was necessary (of the order of 10 mW), R 6G could also be used for all lines studied. The pumping intensity used in most cases was only about 1 mW/cm^2.

In a first experiment the $1s_5$ $(^3P_2) \leftrightarrow 2p_9(^3D_3)$ transition of Ne at $\lambda = 640$ nm was used to create an orientation in the metastable $^3P_2$ state. The Ne sample at 5 mtorr was confined in a sealed spherical pyrex cell, 6 cm in diameter, containing 52 mtorr of He as a buffer gas. The metastable state was populated by an electrodeless discharge at 14 MHz. To modulate the probe polarization, we used either an index-modulated photoelastic crystal (Jobin Yvon) vibrating at 18 KHz, or a $\lambda/4$ plate rotating at 80 Hz.[10] The probe beam was split just before the sample cell to allow differential detection and reduction of the noise due to laser intensity fluctuations. A slight angle between the pump and detection beam allowed overlapping between these two beams. Alternatively, when mirror M of Fig. 1 is small enough, it is possible to use parallel laser beams, as shown in this figure. This is because the metastable orientation can diffuse over relatively large distances, so that one can observe signals from atoms in the "shadow", i.e., not directly interacting with the pump beam. The air pressure change in the laser cavity was measured by using a pressure transducer directly coupled to the X axis of an XY recorder. A Fabry-Perot etalon provided frequency calibration of the scan.

Figure 2(a) shows the recorded orientation spectrum of the 640-nm line. The small amount of ^{22}Ne present in natural neon gives rise to the smaller resonance at higher frequency, thus displaying the isotope shift (not resolved in the Doppler-broadened fluorescene spectrum). The narrow saturation free resonances are superposed on broad backgrounds produced by velocity-changing collisions that partially conserve the orientation. The width of this background is about half the "linear" Doppler width of the fluorescene spectrum. This shows that there is a correlation between the portion of orientation conserved and the amount Δv_z by which the atomic velocity is changed by the collision process.[11]

In this experiment, the atomic levels that have populations modified by optical pumping are nearly degenerate (Zeeman sub levels). These populations

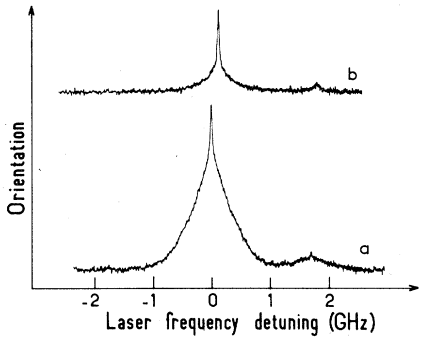

FIG. 2. Recorded spectrum of orientation in velocity selective optical pumping at the 640-nm line in natural neon (91% of ^{20}Ne and 9% of ^{22}Ne) at 5 mtorr with 50 mtorr of ^{4}He. The width of the narrow peaks is about 25 MHz. (a) In the absence of rf field, velocity diffusion of orientation gives a broad background. (b) A magnetic resonance rf field destroys the orientation in the background.

can therefore easily be modified by magnetic resonance techniques. As a demonstration, we performed an experiment by applying a longitudinal magnetic field $\vec{B}_0$ (parallel to $0z$) and a transverse radiofrequency field $\vec{B}_1(t)$, as indicated in Fig. 1. The frequency ν of $\vec{B}_1$ was chosen in order to meet the magnetic resonance condition inside the metastable $^3P_2$ state ($B_0 = 1$ G, $\nu = 2.1$ MHz). Then, in moderate B_1 intensities, the broad background completely disappears, as shown in Fig. 2(b), and only the narrow peaks at the center of the lines remain observable. In a simplified description, the atoms of the velocity group v_z in resonance with the pumping beam are constantly excited out of, and come back to, the metastable state at a rate T_p (T_p is the pumping time). They therefore have a shorter transverse orientation relaxation time T_2 than the atoms which are out of resonance. Those oriented atoms that are Doppler-shifted out of resonance by velocity-changing collisions are therefore depolarized by a weaker rf field than the atoms being pumped.

Many atomic lines can be used to perform this type of experiment. As the excitation of the 640-nm line corresponds to a $J = 2$ $J' = 3$ transition,[12] optical pumping leads to an accumulation of the atoms in the $m_J = +2$ sublevel (this is actually only true at low pressure; see below), thus increasing the absorption of a σ^+ polarized probe beam. On the other hand, if one chooses an atomic resonance line for which $J' = 2$ (or 1), atoms in the $m_J = 2$ sublevel no longer absorb a σ^+ polarized probe. In other words, according to the resonance line used, the same orientation $\langle J_z \rangle$ leads to enhanced absorption, or transparency. To check this result, velocity selective optical pumping was performed in the $^3P_2$ metastable state by using the 595- and 614-nm neon lines ($J' = 2$ for both these lines). The orientation spectra obtained were similar to those of Fig. 2, but with reversed sign.

It is also well known that the optical pumping cycle depends on the collisional depolarization in the upper state. At low pressures, essentially no such collisions occur, and optical pumping is of "Kastler type" (Ref. 13) (as described in the preceding paragraph). On the other hand, when the buffer gas pressure is high enough (several upper-state depolarization collisions occur during the lifetime γ^{-1}), no orientation returns by spontaneous emission to the lower state, and the optical pumping is of "Dehmelt type"[13] (sometimes also called "depopulation pumping"[6]; in this terminology, Kastler-type optical pumping would be a "depopulation-repopulation" pumping). For a $J = 2 \rightarrow J' = 3$ transition, both types lead to opposite orientations in the lower level (accumulation in the $m < 0$ sublevels with a σ^+ Dehmelt-type optical pumping). This prediction was also checked, and Fig. 3 shows the result of the same experiment at 640 nm as in Fig. 2, but with a cell containing Ne at 32 mtorr and He at 320 mtorr. Due to the influence of velocity-changing collisions in this experiment, the situation is actually intermediate between Kastler and Dehmelt pumping conditions. As the mean free time between collisions in the excited state is $\tau_c \lesssim \gamma^{-1}$, the excited atoms in the pumped velocity group undergo velocity-changing collisions with partial conservation of orientation before returning to the metastable state through spontaneous emission. Thus the total orientation of the velocity group in resonance with the pumping beam has an important contribution from a Dehmelt-type process, while the background atoms are mainly oriented by Kastler pumping. This produces a narrow resonance with inverted sign superposed on the background signal.

A particularly interesting case occurs when the orientation of the atomic level is purely nuclear. This can be realized, for example, by studying

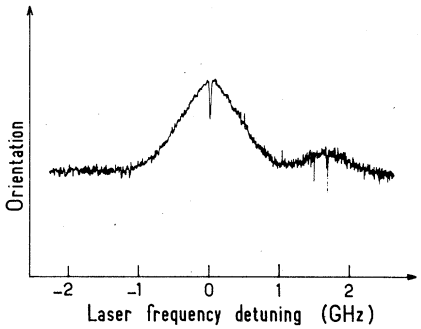

FIG. 3. Orientation spectrum with the 640-mm line in ^{20}Ne and ^{22}Ne at 32 mtorr with 320 mtorr of ^{3}He.

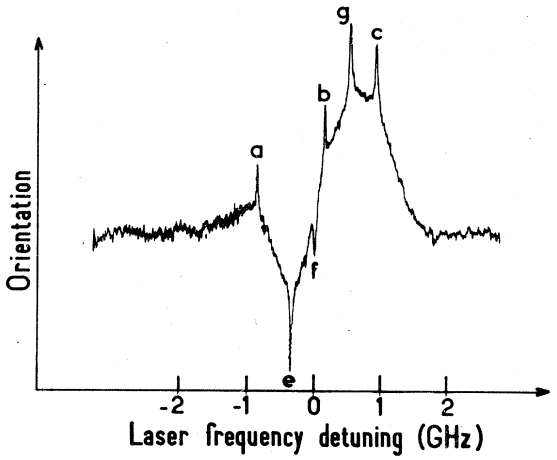

FIG. 4. Orientation spectrum of ^{21}Ne with the 627-nm line. The three hfs components are: (a) $F=3/2 \leftrightarrow 5/2$, and (c) $F=3/2 \leftrightarrow 1/2$; the signals (e), (f), and (g) are crossover resonances.

the $^3P_0$ metastable state of ^{21}Ne. This state has no electronic angular momentum, and the oriented nuclear spin $I=3/2$ is totally insensitive to collisions on time scales of relevance here. In such a case, one could think that no depolarization would take place in velocity-changing collisions and the orientation would diffuse over the total Doppler width. The spectrum would then exhibit only a broad background without any narrow resonance (for the same reason, as pointed out in Ref. 14, velocity selective optical pumping with the resonance line of Na can not be performed by the method discussed here[15]). However, when metastability exchange collisions are present there will be collisional relaxation of the orientation in the $^3P_0$ state. An exchange collision between an oriented metastable atom and a nonoriented atom in the ground state will result in a depolarized metastable atom. If the steady nuclear orientation in the ground state is negligible (e.g., due to depolarization during wall collisions), exchange collisions do introduce a depolarization in velocity-changing collisions, thus allowing for a narrow resonance in the pumped velocity group.

We performed an experiment in the $^3P_0$ state using the 627-nm line ($1s_3$, $J=0 \rightarrow 2p_5$, $J=1$). The resulting spectrum is shown in Fig. 4 (^{21}Ne pressure: 13 mtorr, He pressure: 56 mtorr). The narrow peaks are clearly resolved from the background, as expected with metastability exchange collisions present. The three hfs components ($F=3/2 \rightarrow F'=1/2$, $3/2$, $5/2$) are displayed, as well as the "crossover" resonances at the mean frequency of each pair of hfs components. The relative weights and signs 5:4:−9 of these resonances are determined by the appropriate coupling factors.[8] In fact, as part of the atoms excited to the $2p_5$ state radiate at other wavelengths, the $^3P_0$ state is emptied by absorption, and "Dehmelt pumping" takes place. Pumping at the component $F=3/2 \rightarrow F'=5/2$ produces an orientation which is opposite in sign to that of the other components. The detection process thus restores the same sign for the three main resonances, but a "crossover" resonance between the $F=I=3/2 \rightarrow F'=5/2$ transition and any of the other components necessarily results in a negative resonance, as shown in Fig. 4. The background results from the combined effect of the various components in velocity diffusion. It can be seen in Fig. 4 that this background always has the sign of the nearest narrow peak; it therefore clearly exhibits strong velocity memory effects during atomic collisions. The method presented here may prove useful for the study of metastability exchange in conjunction with velocity-changing and depolarization collisions.

In conclusion, it can be remarked that, in general, whenever the optical excitation rate exceeds any of the relaxation rates for one velocity class of atoms in a gas, Doppler-free signals can be obtained. The velocity selective optical pumping method saturates depolarization or velocity-changing collision processes, in experimental conditions where no saturation of spontaneous decay occurs ($T_p \gg \gamma^{-1}$). It seems perfectly possible that part of the signals observed in saturated-absorption-type experiments (high intensities, nonexpanded beam) may be due to velocity selective optical pumping effects. This is especially true in the study of molecular transitions with low oscillator strengths, but still short enough excited-state lifetime, and when the sample is contained in a cell. In this article, an experiment is described where optical pumping is isolated, so that no significant optical coherences are present and no optical saturation occurs. The expanded pumping and narrow probe beams insure spatially homogeneous conditions. The detection of perfectly well-defined observables as well as the possibility of selective action by magnetic resonance considerably simplifies the calculations and the interpretation of the collision effects. However, since a necessary condition for effective optical pumping is $T_p \lesssim T_1$ (orientation relaxation time), the condition $\gamma^{-1} \ll T_p \lesssim T_1$ must be met. The main restriction of the method presented here is therefore that the lifetime of one of the atomic levels involved in the optical transition has to be long compared to the lifetime of the other level.

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[7]With a linear pump polarization, the observable created would be alignment components, e.g., $\langle 3J_z^2 - J(J+1)\rangle$, etc.

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[10]In another version of the experiment, both the pumping and probe polarization are modulated, and the sum or difference frequency is monitored. This strongly reduces parasitic signals (spurious reflections, etc.).

[11]We also detected the circular birefringence induced in velocity selective orientation. Probing with linear polarization and modulating the circular polarization $\sigma^{\pm}$ of the pumping beam yields a modulated Faraday rotation of the probe polarization plane. This was detected through polarizers crossed at 45°, and the recorded spectra showed the corresponding birefringence resonances: a narrow dispersion curve superposed on a broad dispersion-shaped background.

[12]An advantage of this transtion is that the atoms necessarily return to the $^3P_2$ state by spontaneous emission ("closed" system).

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[14]M. Elbel, H. Hühnermann, Th. Meier, and W. B. Schneider, Z. Phys. A 275, 339 (1975).

[15]For the ground state of Na atoms, the depolarization collision cross section is much smaller than the velocity-changing cross section. In order to obtain velocity selective optical pumping, one must either have a short interaction time T_i (focused pump beam, low gas pressure) or perform a transient experiment [T. W. Hänsch, I. S. Shahin, and A. L. Schawlow, Phys. Rev. Lett. 27, 707 (1971)]. Of course, narrow peaks could also be observed in the presence of optical saturation.