PHYSICAL REVIEW A VOLUME 34, NUMBER 3 SEPTEMBER 1986

Complete Doppler coverage in laser optical pumping by wall-induced velocity-changing collisions

C. H. Holbrow,* A. P. Ghosh, D. Heinzen, X. Zhu,† W. W. Quivers, Jr., G. Shimkaveg,
P. G. Pappas,‡ J. E. Thomas, and M. S. Feld
George R. Harrison Spectroscopy Laboratory and Department of Physics, Massachusetts Institute of Technology,
Cambridge, Massachusetts 02139

(Received 25 February 1985; revised manuscript received 9 April 1986)

Enhanced Doppler coverage has been observed in the laser-induced optical pumping of low-density rubidium vapor in the absence of any buffer gas. Transient and steady-state measurements show that this enhancement arises from velocity-changing collisions (VCC's) of rubidium atoms with the walls of the cell containing the vapor. Wall-induced VCC's offer a possible mechanism for increasing the efficiency with which atomic and nuclear polarization may be produced with lasers.

Laser optical pumping is a promising way to produce polarized vapors for fusion fuels,[1] for charge- and spin-exchange media,[2] and as sources and targets for experiments in nuclear physics.[3] This process produces nuclear orientation by transferring angular momentum from an incident beam of light to the atomic ground state via a succession of resonant absorptions and spontaneous emissions.[4] In appropriate circumstances a dye laser of moderate power can polarize dense samples.[3,5]

Unfortunately, at vapor pressures commonly used the effectiveness of laser optical pumping may be limited because monochromatic laser radiation interacts strongly only with those atoms of the Maxwellian velocity distribution that are Doppler shifted into resonance. Therefore, only a small fraction of the atoms—the ratio of the homogeneous linewidth to the width of the Doppler distribution—can be optically pumped.

Several ways to increase Doppler coverage are known. Power broadening is effective,[6] but uses laser power inefficiently.[5] Multiple lasers have been used, but such arrangements are awkward as well as expensive.[7] Small amounts of inert buffer gas added to the sample vapor have been used to produce velocity-changing collisions (VCC's) that rapidly redistribute the atomic velocities without reequilibrating the populations of the atomic levels. In this way VCC's permit optical pumping of the entire Doppler distribution.[5] The technique makes efficient use of laser power, but for some applications the presence of buffer gas is undesirable. A multimode laser can be used to pump over a Doppler distribution. In one such case,[8] by modulating the laser's thin etalon the mode distribution has been matched to the Doppler width to achieve efficient pumping.

This paper describes a way to achieve Doppler coverage using VCC's but without using a buffer gas. We have used collisions with the walls of the vapor cell to improve Doppler coverage. Our measurements of Lamb-dip saturation spectra of optically pumped rubidium vapor show that wall-induced VCC's strongly enhance Doppler coverage even when the optically pumped atoms undergo only twenty or so collisions with the walls before relaxation.

To study Doppler coverage produced by wall-induced changes of velocity we used a standard Lamb-dip configuration in which a cell of rubidium vapor was illuminated with a chopped pump beam and a counterpropagating probe beam from the same laser. The cell, a Pyrex cylinder 2 cm in diameter and 3 cm long, was placed in a keeper magnetic field of ~ 3 G parallel to the cylinder's axis. The laser, a Coherent 599-21 frequency-stabilized dye laser, was tuned to the rubidium D_1 resonances. The pump light was a beam of TM_{00} mode radiation of intensity 130 $\mu\mathrm{W/cm^2}$ directed along the axis of the cell. The probe beam intensity was kept small, ~ 0.2 $\mu\mathrm{W/cm^2}$, to avoid unwanted optical pumping effects. The pump beam diameter was larger than that of the probe and, except where indicated, their axes coincided. Experiments were run both with linearly polarized and circularly polarized pump light. In each case the probe had the same polarization direction as the pump light. Phase-sensitive detection was used to monitor variations in transmission of the probe beam as the laser was tuned through the D_1 resonances.

The presence of VCC's of some sort is evident in the Lamb-dip spectrum of the D_1 hyperfine structure of $^{85}\mathrm{Rb}$ shown in Fig. 1. In the absence of VCC's only the three sharp peaks should appear as the laser is tuned across the resonances. The left and right peaks arise, respectively, from the $S_{1/2}F=3\rightarrow P_{1/2}F=2$ and $S_{1/2}F=3\rightarrow P_{1/2}F=3$ transitions when both pump and probe are resonant with the same atomic velocity group, as can only occur for atoms at zero velocity parallel to the beams, i.e., atoms at the center of the Doppler profile. The middle peak, a crossover resonance, is a familiar Lamb-dip artifact. However, the pronounced pedestal on which these sharp peaks sit reveals that after atoms of velocity v were pumped they came into resonance with the counterpropagating probe beam, and so must have undergone a shift in velocity to $-v$.

Our experimental setup was designed to eliminate all sources of VCC's except the walls. No buffer gas was introduced. Before loading rubidium into the cell, it was baked out at 300 °C at 10^{-6} Torr for an hour. When in use the system was pumped through a liquid-nitrogen cold

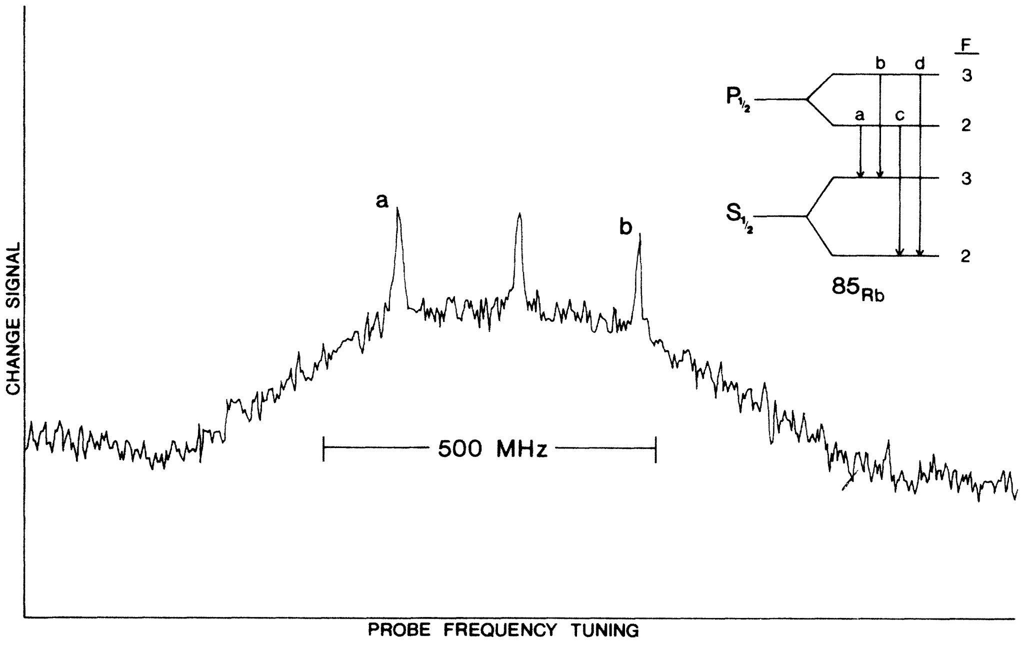

FIG. 1. Lamb-dip signal for ^{85}Rb D_1 hyperfine transitions. The peaks labeled *a* and *b* correspond to the transitions in the inset.

trap with a small oil diffusion pump. Background pressure was at most a few μ Torr. Rubidium vapor pressure was controlled with a heater and was kept low enough so that the mean-free-path of a rubidium atom was always considerably more than the dimensions of the cell. Nevertheless, VCC pedestals appear prominently over a range of vapor densities from 5.4×10^{9} cm^{-3} (22 °C) to 1.2×10^{13} cm^{-3} (120 °C), and in all lines of the spectra of both the D_1 and D_2 lines of both ^{85}Rb and ^{87}Rb and for all polarizations of the pump beam. The only possible source of VCC's is the cell walls.

To show more clearly that it is the walls that enhance Doppler coverage, the pump and probe beams were kept small (respectively, 5.3 mm and 2.1 mm in diameter) relative to the 20-mm diameter of the cell, and were kept away from the walls. Under these circumstances there is an annular volume with which the laser pump beam does not interact.

If, as we assert, pumped atoms drift to the walls and undergo VCC's without relaxation, the annular region should acquire a population of optically pumped atoms with velocities that Doppler shift them into resonance with the probe beam. Such atoms were detected by separating the pump and probe beams by several millimeters. In this way the probe beam saw only atoms in the unpumped volume. Nevertheless, the Lamb-dip spectra continued to exhibit pronounced pedestals, although no sharp peaks. The presence of the pedestals proves that VCC's occurring at the walls produce in the unpumped volume a population of optically pumped atoms resonant with the probe beam.

The effect of the walls is further confirmed by measurements of transient decay of the pumped atoms. Using a boxcar integrator and a chopper that gave a 3 μs rise time, we measured the rate at which the variation in the probe

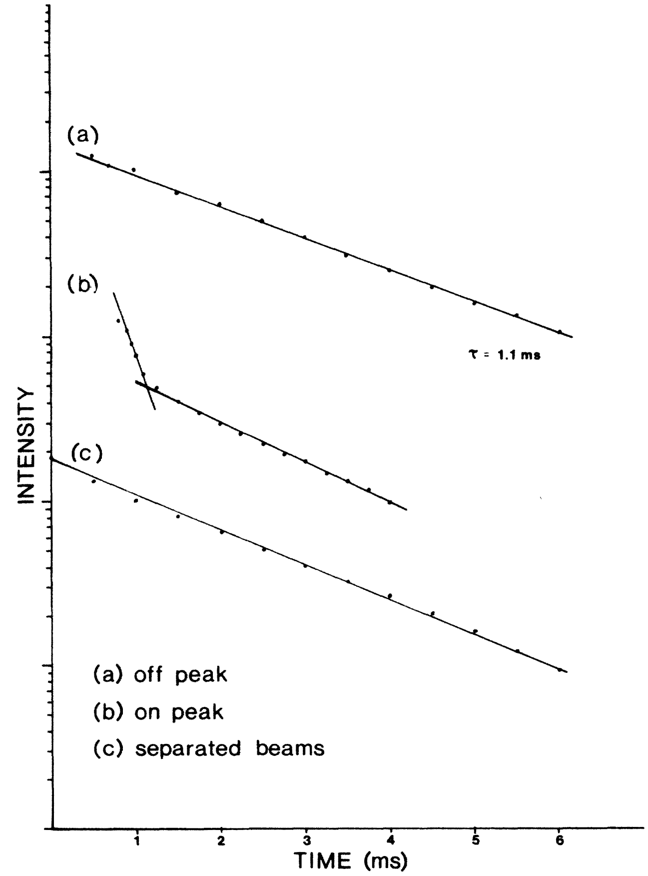

FIG. 2. Transient decays of the change signals of the probe. The pump beam is turned off at $t=0$. The probe laser is tuned to (a) the pedestal only and (b) to a Lamb-dip peak. In (c) the probe beam is separated spatially from the pump beam.

signal decayed after the pump was turned off. Figure 2 shows experimental results for different situations. In Fig. 2(b) the probe is tuned to the middle of the Lamb-dip peak and in Fig. 2(a) the probe is tuned off the peak but still on the pedestal. In both cases the pump and probe beams are coaxial. Figure 2(c) shows the transient decay for an experiment in which the probe beam was displaced parallel to the pump beam.

The semilog plots of the signals show clearly that when the probe is tuned to the peak of the saturation resonance there is a fast exponential decay followed by a slow one with a time constant of 1.1 ms. The apparent value of the time of the fast decay arises from the gate width of the boxcar integrator and not the behavior of the atoms. From a separate measurement using an oscilloscope the time constant of the fast decay was found to be ~ 10 μs. With the probe tuned to the pedestal but off the Lamb dip there is only a slow decay time of 1.1 ms.

These observations are all accounted for by the effects of wall-induced VCC's. The fast decay should arise from the drift of pumped atoms out of the probe beam. This would occur in a time of the order of the beam radius ($\sim$2.65 mm) divided by the atomic velocity ($\sim$300 m/s), which is ~ 9 μs, close to the observed value of 10 μs. The failure to detect fast decay, i.e., a Lamb dip, in the unpumped volume is attributable to the decrease in density of the corresponding atoms as they diffuse out from the pumped region toward the wall, and to limits on the sensitivity of our detection of the fast transient.

The slow decay is also consistent with the picture of wall-induced VCC's. Since a pedestal is the result of a population of pumped atoms redistributed with respect to velocity by collisions with the walls and filling the entire volume of the cell, the pedestal decays only as the atoms relax from their pumped state. Presumably relaxation takes place at the walls but, of course, it must take place less frequently than collisions with the walls in order for pedestals to be present at all. The time for an atom to drift from the probe beam to the wall and back is ~ 50 μs. Therefore, the observed relaxation time of 1.1 ms implies an atom collides with the wall more than 20 times before it relaxes from the pumped state. Such collisions are well known for rubidium.[9] With special wall coatings there can be tens of thousands of such collisions before relaxation occurs. Although no special wall coating was used in our Pyrex cell, its walls probably had adsorbed oil from the diffusion pump.

Several improvements of the technique described above are needed in order to maximize polarization of the atoms. First, provision has to be made to keep atoms from becoming inaccessible to the laser field because of hyperfine or F pumping. A second laser or RF ground-state mixing is needed to keep atoms from being lost by pumping into the $F=3$ or $F=2$ levels of the ground state (Fig. 1 inset). Second, by increasing the collisional relaxation time one can both reduce the intensity of laser light required to saturate the optical pumping and increase Doppler coverage. Bouchiat[9] has already shown that some wall coatings can increase relaxation times for rubidium by 10^3. Finally, one can irradiate the entire cross section of the cell with pump light, and thereby access the maximum possible numbers of atoms. With these modifications wall-induced VCC's can usefully enhance the nuclear polarization obtainable by laser optical pumping.

This work was supported by the National Science Foundation and the Colgate University Research Council.

---

[*] Permanent address: Department of Physics and Astronomy, Colgate University, Hamilton, NY 13346.

[†] Permanent address: Wuhan Institute of Physics, Academia Sinica, Wuhan, Hubei, The People's Republic of China.

[‡] Present address: AT&T Bell Laboratories, Summit, NJ 07901.

[1] R. Kulsrud, H. Furth, E. Valeo, and M. Goldhaber, Phys. Rev. Lett. **49**, 1248 (1982); D. E. Murnick, App. Phys. Lett. **42**, 544 (1983).

[2] N. D. Bhaskar, W. Happer, and T. McClelland, Phys. Rev. Lett. **49**, 25 (1982).

[3] G. J. Witteveen, Nucl. Instrum. Methods **158**, 57 (1979); L. W. Anderson, Nucl. Instrum. Methods **167**, 363 (1979); L. W. Anderson, in *Lasers in Nuclear Physics,* edited by C. E. Bemis, Jr. and H. K. Carter (Harwood, New York, 1982), p. 313, and other articles therein; G. Shimkaveg, W. W. Quivers, Jr., R. R. Dasari, C. H. Holbrow, P. G. Pappas, M. A. Attili, J. E. Thomas, D. E. Murnick, and M. S. Feld, Phys. Rev. Lett. **53**, 2230 (1984); F. P. Calaprice, W. Happer, D. F. Schreiber, M. M. Lowry, E. Miron, and X. Zeng, *ibid.* **54**, 174 (1985).

[4] A. Kastler, J. Phys. Rad. **11**, 255 (1950); B. Cagnac, Ann. Phys. (Paris) **6**, 467 (1961).

[5] P. G. Pappas, R. A. Forber, W. W. Quivers, Jr., R. R. Dasari, M. S. Feld, and D. E. Murnick, Phys. Rev. Lett. **47**, 236 (1981); J. E. Thomas and M. S. Feld, *Lasers in Nuclear Physics,* Ref. 3, p. 339.

[6] W. D. Cornelius, D. J. Taylor, R. L. York, and E. A. Hinds, Phys. Rev. Lett. **49**, 870 (1982).

[7] E. A. Hinds, W. D. Cornelius, and R. L. York, Nucl. Instrum. Methods **189**, 599 (1981).

[8] E. W. Weber and H. Vogt, Phys. Lett. **A103**, 327 (1984).

[9] M. A. Bouchiat and J. Brossel, Phys. Rev. **147**, 41 (1966).