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Concentration of atomic population in any single-ground-state magnetic sublevel in alkali-metal vapors

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A scheme for concentrating a large fraction (~100%) of the alkali-metal atoms in an atomic vapor in *any single* magnetic sublevel of the ground state is developed. This is experimentally demonstrated in the ^{87}Rb vapor. The atomic vapor is optically pumped to a high degree of spin polarization after which two radio-frequency π pulses are applied to transfer the atomic population from the (2,2) or (2,−2) (F,M_F) state to the (2,0) sublevel of the ground state. The resulting distribution is diagnosed using a microwave field tuned to the hyperfine transition (6834 MHz).

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Optical pumping of alkali-metal beams and vapors continues to be a subject of considerable interest for understanding fundamental physics, as well as having a wide range of applications such as polarized beams and targets for spin-dependent scattering studies in both atomic and nuclear physics, polarized noble gases, and heavy atoms for studying fundamental symmetries (searches for permanent electric dipole moments in atoms), surface studies, magnetometers, and optically pumped frequency standards [1–5]. Spin-exchange optical pumping [6] has has been used extensively for many fundamental studies [7]. In alkali-metal vapors, using circularly polarized $D1$ resonance light from a tunable laser, nearly all of the atoms can be easily transferred to either $|F\ M_F=F\rangle$ or $|F\ M_F=-F\rangle$ magnetic sublevels of the ground state (100% spin polarization), where $F=I+\frac{1}{2}$ and I is the nuclear spin. By selectively exciting one of the two ground hyperfine levels, a large fraction of the atomic population can also be pumped to one of the two ground hyperfine levels $F=I+\frac{1}{2}$ or $F=I-\frac{1}{2}$ (hyperfine polarization); however, the population is equally distributed among *all* the $(2F+1)$ magnetic sublevels. These types of nonthermal population distributions play a key role in many optically pumped atomic devices (e.g., the rubidium gas cell atomic frequency standard and magnetometer).

An interesting question regarding population distributions is: *Can one concentrate all of the alkali-metal atoms in a vapor in any one of the 2(2I+1) ground-state magnetic sublevels?* Such an ensemble could be of fundamental interest; e.g., for scattering studies in atomic and nuclear physics and relaxations of various types of multipole distributions. Dreves *et al.* [8], using optical pumping with circularly polarized light from a tunable laser together with microwave and radio-frequency (rf) fields, demonstrated selective population concentration in a single magnetic sublevel in a Na *atomic beam.* Avila *et al.* [9] have demonstrated, in a Cs atomic beam, the concentration of population in one of the two $M_F=0$ sublevels of the ground state. They use two tunable lasers and utilize the optical selection rules. Currently there is considerable interest in developing optically pumped cesium-beam frequency standards with all the Cs atoms concentrated in one of the two $M_F=0$ ground sublevels. In an atomic beam there are no collisional relaxations; however, in an atomic vapor, relaxations tend to rapidly equalize the population among the magnetic sublevels. Therefore, producing a desired population distribution in an atomic vapor is considerably more complex than in the atomic beam.

We have developed a scheme for concentrating a large fraction of the ground-state atoms in an alkali-metal *atomic vapor* in *any one* of the ground magnetic sublevels. We use optical pumping with circularly polarized light in conjunction with pulsed rf. Diagnostics of the resulting population distribution are performed using a microwave field. We have experimentally demonstrated the concentration of a large fraction of the ^{87}Rb atoms in one of the two $M_F=0$ magnetic sublevels of the ground state. In this paper we present the pertinent details of our experiment together with the results.

The experimental arrangement is shown in Fig. 1. A sealed cylindrical Pyrex bulb containing several milligrams of ^{87}Rb and various buffer gases (10 Torr N_2 and 100 Torr He measured at 25 °C) is placed at the center of a Helmholtz coil assembly (three mutually orthogonal pairs of Helmholtz coils). A uniform dc magnetic field in the range of 1–30 G is produced along the Z direction. The X and Y coils are used to cancel the residual magnetic field in the transverse plane. A single-mode $Al_xGa_{1-x}As$ diode laser of 5 mW output power and tuned to the $D1$ transition (794.7 nm) is used for optical pumping. 10 Torr of N_2 is used to quench the laser-excited resonance fluorescence, which would otherwise degrade the atomic polarization [10]. 100 Torr of He serves to slow the diffusion of the oriented atoms to the glass wall where they undergo collisional depolarization. Buffer-gas pressure is chosen so as to keep the pressure-broadened absorption linewidth substantially smaller than the ground-state hyperfine splitting (6834 MHz). To produce a high degree of orientation in the vapor the frequency of the pump laser is tuned midway between the two ground-state absorption lines. This allows for approximately equal excitation of both the ground hyperfine levels. With circularly polarized light nearly all

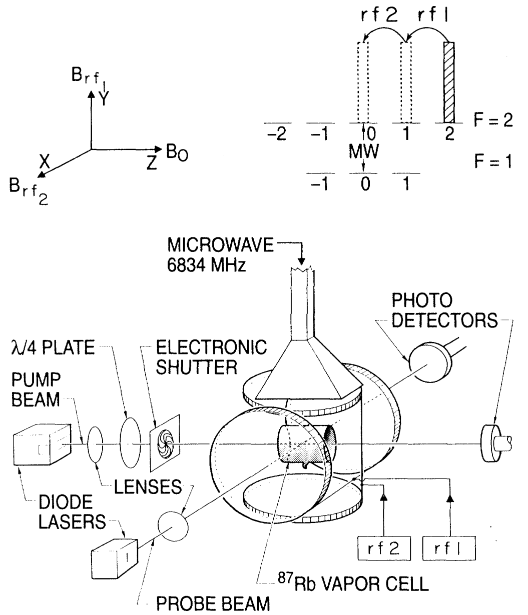

FIG. 1. A schematic of the experimental apparatus. The Rb cell is maintained at 50 °C. The sequence of the pulses to transfer the atomic population from (2,2) to (2,0) (F,M_F) is also shown.

of the ground-state atoms are optically pumped into either the (2,2) or (2,−2) (F,M_F) sublevels, depending on the handedness of the circular polarization. To produce a large hyperfine polarization the laser is linearly polarized and tuned to excite the atoms in one of the two ground hyperfine levels. Two pairs of circular coils are used to generate two independent rf magnetic fields in the X and Y directions. A horn supplies the diagnostic microwave magnetic field (6834 MHz). The horn is oriented so that the microwave magnetic field is parallel to the Z axis to excite $\Delta M_F=0$ transitions between the ground-state hyperfine levels. The rf and microwave fields are produced using frequency synthesizers and frequency multiplier chains.

With circularly polarized light nearly 100% spin polarization is easily accomplished. The optical pumping rate ($\sim 3000\ s^{-1}$) is much larger than the relaxation rates (150–200 s^{-1}). The relative population distribution among the magnetic sublevels is easily measured using the technique of rf Zeeman spectroscopy. At low magnetic fields (a few gauss) the six Zeeman (magnetic resonance) transitions (four for $F=2$ and two for $F=1$) are essentially degenerate. However, for a magnetic field of about 20 G all the transitions are completely resolved. The rf transition frequencies can be easily calculated using the Briet-Rabi formula [11]. In our experiment the rf resonance frequency is 15–16 MHz. The rf fields induce transitions between the adjacent magnetic sublevels ($\Delta F=0$, $\Delta M_F=\pm 1$). This results in the reduced transmission of the pump beam. The magnetic resonance spectrum is dominated by a single line corresponding to the (2,2) to (2,1) [or (2,−2) to (2,−1)] transition; all other transitions are practically zero—indicating a complete concentration of the atomic population in the highest angular momentum state.

Transferring the atomic population from the initial (2,2) [or (2,−2)] state to any other magnetic sublevel with a minimal loss is a challenging problem. We use rf pulses to transfer the atomic population. A virtually complete inversion of the population between the two adjacent Zeeman levels [e.g., (2,2) and (2,1)] can be achieved by applying a pulse of rf [rf1] tuned to the Zeeman transition frequency. The requirement on the rf pulse is that $\omega_1\tau=\pi$, where ω_1 is the Rabi frequency and τ is the duration of the pulse. In the NMR terminology this is referred to as a π pulse [12]. Since initially all the atoms are concentrated in the (2,2) level, application of such a pulse leads to a complete transfer of this population to the (2,1) level. Immediately after this a second π pulse (rf2) tuned to the (2,1)-(2,0) transition frequency transfers the large atomic population from the (2,1) to the (2,0) sublevel. These operations are done in rapid succession to minimize the loss due to various relaxations taking place in the vapor. Though the rf couples only the two adjacent levels, spin-exchange collisions and optical pumping connect all of the $2(2I+1)$ magnetic levels. These pulses are applied with the strong pumping light turned off (using an electronic shutter), which otherwise would tend to push the atomic population to the high angular momentum state. At the end of the two independent rf π pulses a large fraction of the atoms, which were initially concentrated in the (2,2) sublevel, is thus transferred to the (2,0) sublevel.

The population of the (2,0) level is probed using another single-mode $Al_xGa_{1-x}As$ diode laser operating at 780 nm ($D2$ transition) in conjunction with the microwave field tuned to the (2,0)-(1,0) transition ($\sim$6834 MHz). The probe beam is adjusted to overlap with the pump beam. The probe intensity is kept very low (weak probe) and is left on continuously, even during the pump phase. The probe beam is *linearly polarized and is transverse to the pump beam.* It is tuned to optically excite only *one* of the two ground-state hyperfine levels. With 100 Torr of He in the cell, the ground-state hyperfine levels are well resolved (pressure-broadened absorption linewidth is $\sim$2.8 GHz), but the excited-state hyperfine levels are completely smeared. The microwave field is pulsed and applied immediately after the second rf π pulse. The pulse sequences are also shown in Fig. 1. The resonant microwave induces a coherence between the (2,0) and (1,0) levels, resulting in the well-known Rabi oscillations [13]. The transmission of the probe beam undergoes corresponding oscillations, with the Rabi frequency of the oscillations proportional to the microwave magnetic field. The initial amplitude of microwave-induced oscillatory signal is directly proportional to the difference in population between the (2,0) and (1,0) levels. The amplitude of these oscillations decays rapidly due to various relaxation processes (diffusion and spin-exchange collision) and also due to the inhomogeneities of the microwave magnetic field. The experimentally observed signal is shown in Fig. 2. The duration of the microwave pulse is such as to enable one to observe several oscillations. The pumping sequence is repeated and the signal averaged.

The microwave-induced signal is found to be critically

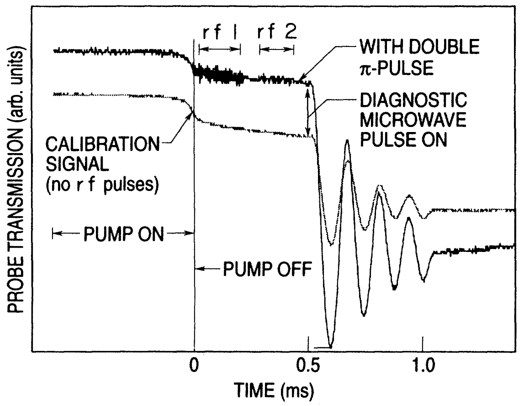

FIG. 2. Microwave-induced signal: Comparison of the double π pulse transfer scheme with the conventional hyperfine polarization method. In the former case, the pump beam is circularly polarized and tuned to produce $\sim 100\%$ orientation in the atomic vapor. In the latter the pump beam is linearly polarized and tuned to produce a large hyperfine polarization-on rf pulse applied. The microwave field is used only for diagnostics.

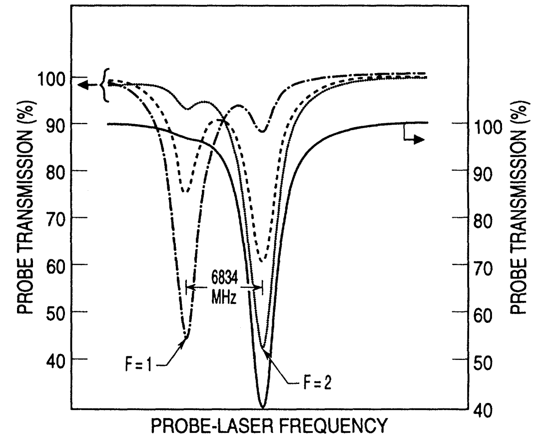

FIG. 3. Absorption profiles of the atomic vapor for various pumping conditions. This is used for calibrating the microwave-induced signals shown in Fig. 2 in terms of Δ, the fractional population difference between (2,0) and (1,0) levels. The dotted line and broken-dotted line, pump linearly polarized and tuned to excite $F=1$ and $F=2$, respectively; dashed line, pump off—thermal population distribution; solid line, pump circularly polarized and tuned to $F=1$. The fractional population of $F=2$ is ~ 1. $[\mathrm{Rb}]\sigma L=0.92$. For the solid line, refer to the axis on the right (probe transmission, percent).

dependent on the two rf π pulses. The microwave signal occurs only when both π pulses are applied in the correct sequence, frequencies tuned to the appropriate transitions, and pulse widths suitably adjusted. In order to assess the efficiency of the population transfer we need a suitable calibration of the microwave-induced signal in terms of Δ, the fractional population difference between the (2,0) and (1,0) levels. The calibration is done in the following manner. A large hyperfine polarization is produced using a linearly polarized pump tuned to excite either the $F=1$ or $F=2$ level. The atomic population in the (2,0) level is probed using the microwave field—essentially the same scheme described above but *without* the rf π pulses. The microwave-induced signal is directly proportional to $\Delta=f_2/5-(1-f_2)/3$, and f_2 is the fraction of the atoms in the $F=2$ level. f_2 is determined accurately from the absorption scans. The absorption profile of the optically pumped vapor obtained by scanning the frequency of the probe beam is shown in Fig. 3. In the absence of the pump beam, the absorption profile (dashed line) corresponds to the unpolarized vapor with population distributed in direct proportion to the statistical weights ($\frac{5}{8}$th and $\frac{3}{8}$th, respectively, in the $F=2$ and $F=1$ levels)—thermal distribution. When the pump beam is circularly polarized and tuned to the $F=1$ level, essentially *all* of the atoms are pumped into the $F=2$ level—absorption of the probe beam at $F=1$ is ~ 0 (solid line). This provides a calibration of the optical density of the vapor relative to the fraction of the atoms in the two hyperfine levels. The transmittance of the pumped vapor at $F=2$ is 0.4. This corresponds to $f_2[\mathrm{Rb}]\sigma L=0.92$; [Rb] is the total number density of the Rb atoms ($f_2=1$ for solid line), σ the optical absorption cross section for the linearly polarized probe laser photons, and L the optical path length of the probe beam. From the transmittances and knowing $[\mathrm{Rb}]\sigma L$, we calculate f_2 for the hyperfine polarized vapor produced by the linearly polarized pump beam. With the linearly polarized pump tuned to $F=2$ (broken-dotted line) only 90% of the ground-state atoms are transferred into the $F=1$ level—with equal distribution among the three magnetic sublevels. The dotted line corresponds to the linearly polarized pump tuned to $F=1$; over 95% of the atoms are transferred to the $F=2$ level, with equal distribution among the five magnetic sublevels. From Fig. 3, $\Delta_{\mathrm{cal}}=0.28$ for the case where the linearly polarized pump is tuned to $F=2$. The calibration signal is also shown in Fig. 2. Comparing the two microwave-induced signals in Fig. 2 we find that Δ_{tran}, corresponding to the double rf pulse scheme, is 0.7. $\Delta=1$ corresponds to all of the atoms in the (2,0) level, a situation that would arise with the double rf π pulse scheme transfer when all the atoms from the (2,2) level are moved into the (2,0) level without *any loss* during the transfer. In other words, although we start with a nearly complete concentration of the atoms in the (2,2) level in our experiment, the pulse transfer scheme moves only 70% of the atoms to the (2,0) state, a 30% loss of atoms during the transfer.

A number of factors contribute to this loss during transfer. The inhomogeneities in the rf magnetic-field strength across the volume of the cell will cause a temporal spread in the π pulse widths for complete population inversion. rf magnetic fields are produced by pairs of circular coils with radii only three times as large as that of the cell. This choice was dictated by our need for high rf field strengths at the center. Inhomogeneities in the static magnetic field will also spread the magnetic resonance transition frequencies. This will not be significant for two reasons. Our Helmholtz coils are designed to produce minimum field inhomogeneities. We have taken considerable caution in using nonmagnetic materials in the vicinity of the cell. The rf lines are substantially power broadened due to the high rf power applied for the π pulses. Collisional relaxations will also contribute to the transfer loss. Typically the π pulses are applied for 200μs—the loss due to collisional relaxations will be negligibly small during this period. The damping of the transient nutation [14] of the rf-induced signal is substantially faster than that which can be accounted for by collisional relaxation, which confirms the presence of large inhomogeneities in the rf fields. We believe the major cause for the loss of population during the transfer is due to the inhomogeneities in the rf magnetic-field strengths inside the cell. The rf field can be made more uniform across the volume of the cell with circular coils of much larger diameter. rf amplifiers can be used to increase the field strength at the center of the cell. Together these will ensure a loss-free transfer of the population.

We have demonstrated a scheme for concentrating atomic population in Rb vapor in one of the two $M_F=0$ magnetic sublevels of the ground state, starting with the ensemble in the initial state of nearly 100% orientation. We use rf π pulses to move the atomic population. The rf pulse scheme necessitates magnetic fields of tens of gauss in order that the two rf transition frequencies be different. This type of transfer scheme would work equally well with microwave π pulses. In our experiments we observe that the microwave transitions between $F=1$ and $F=2$ are very well resolved, even for low magnetic fields (500 mG). Two microwave π pulses can be used to move atoms from (2,2) to (2,0) via (1,1) and a single π pulse to move from (2,2) to (1,1). Starting with a highly oriented vapor, one can move the atomic population to any magnetic sublevel of the ground state.

These types of distributions could potentially be of considerable interest in scattering studies in both atomic and nuclear physics. Another area of impact is the Rb gas-cell frequency standard. The short-term frequency stability of these standards is approximately 10^{-11} for 1 s. In these standards optical pumping is accomplished using resonance lamps, and consequently Δ, the fractional population imbalance between (2,0) and (1,0), is very small ($<1\%$). With our scheme, nearly a 100% population imbalance can be produced, thereby offering the potential for greatly improving the frequency stability of these standards.

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