Push-Pull Optical Pumping of Pure Superposition States

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A new optical pumping method, "push-pull pumping," can produce very nearly pure, coherent superposition states between the initial and the final sublevels of the important field-independent 0-0 clock resonance of alkali-metal atoms. The key requirement for push-pull pumping is the use of $D1$ resonant light which alternates between left and right circular polarization at the Bohr frequency of the state. The new pumping method works for a wide range of conditions, including atomic beams with almost no collisions, and atoms in buffer gases with pressures of many atmospheres.

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In 1961, Bell and Bloom [1] first showed that it is possible to produce a coherence between the Zeeman sublevels of alkali-metal atoms by pumping the atoms with light modulated at the Zeeman resonance frequencies. In 1976, Alzetta *et al.* [2] showed that different longitudinal modes of a laser, with frequencies differing by the hyperfine frequencies of alkali-metal atoms, could induce analogous coherences between the hyperfine sublevels. Resonances induced by modulated light are often called "coherent population trapping" (CPT) resonances [3]. They are of considerable current interest for potential applications in quantum computation [4], for maser [5] and laser systems [6], and for atomic clocks [7,8].

In all prior work we know of, a relatively small fraction of the atoms can be pumped into a superposition state. The new "push-pull" pumping method described here makes it possible to pump almost all alkali-metal atoms into the important 0-0 superposition state, which is widely used in atomic clocks because its resonance frequency is insensitive to magnetic fields. In our preliminary experiments to test these ideas, the CPT resonances observed with push-pull pumping were 1 to 2 orders of magnitude larger than those observed under the same conditions with conventional pumping. Push-pull pumping may substantially improve the performance of atomic clocks.

To simplify the discussion, consider pulses of $D1$ light, propagating along the direction of the magnetic field and separated by $T_{00}/2$, half of the period T_{00} of the 0-0 transition. Pulses of right circular polarization (RCP) alternate with pulses of left circular polarization (LCP). We also assume for simplicity that the pulse widths are much shorter than $T_{00}/2$.

Let the alkali-metal atoms have a half-integer nuclear spin quantum number I, and let a small magnetic field B define the z axis of a coordinate system. We denote the energy sublevels of the ground state by $|fm\rangle$. The total spin angular momentum operator $\mathbf{F} = \mathbf{S} + \mathbf{I}$ is the sum of the electron and nuclear spin operators $\mathbf{S}$ and $\mathbf{I}$. The azimuthal quantum number m is defined by $F_z|fm\rangle = m|fm\rangle$. For small magnetic fields, the total angular momentum quantum number f is defined by $\mathbf{F}\cdot\mathbf{F}|fm\rangle \approx f(f+1)|fm\rangle$ where $f = I + 1/2 = a$ (for the upper hyperfine multiplet) or $f = I - 1/2 = b$ (for the lower hyperfine multiplet).

Let the 0-0 superposition state be described at time t by

$$|\psi\rangle = \frac{|a0\rangle e^{-iE_{a0}t/\hbar} + |b0\rangle e^{-iE_{b0}t/\hbar}}{\sqrt{2}}. \quad (1)$$

Here E_{a0} and E_{b0} are the energies of the basis states $|a0\rangle$ and $|b0\rangle$. The phases of the basis states can be chosen such that $\langle a0|S_z|b0\rangle = 1/2$. Then the expectation value of the longitudinal electron spin is

$$\langle S_z\rangle = \langle\psi|S_z|\psi\rangle = \tfrac{1}{2}\cos\omega_{00}t. \quad (2)$$

The Bohr frequency is $\omega_{00} = (E_{a0} - E_{b0})/\hbar = 2\pi/T_{00}$.

In the high-pressure limit, the probability for a spin-polarized alkali-metal atom to be excited by a light pulse is proportional to $(1 - 2\mathbf{s}\cdot\langle\mathbf{S}\rangle)$, where $\mathbf{s}$ is the expectation value of the photon spin [9]. Then the relative probabilities p_+ and p_- of absorbing RCP and LCP pulses by the superposition state of Eq. (1) are

$$p_+ = \sin^2\frac{\omega_{00}t}{2} \quad \text{and} \quad p_- = \cos^2\frac{\omega_{00}t}{2}. \quad (3)$$

As sketched in Fig. 1, we assume that RCP pulses hit the atoms at the times $t = 0$, T_{00}, $2T_{00}$, etc., when $p_+ = 0$. LCP pulses hit the atoms at the times $t = \frac{1}{2}T_{00}$, $\frac{3}{2}T_{00}$, $\frac{5}{2}T_{00}$, etc., when $p_- = 0$. Atoms in the superposition state $|\psi\rangle$ of Eq. (1) will absorb very little light from pulses of either polarization. Atoms in any other state will be excited by the pulse train. No matter what the initial state of the atoms is, nearly all of them will eventually be pumped into the superposition state of Eq. (1), the "dark state" of the system.

Push-pull pumping can be modeled quantitatively by including the following physics: (a) The evolution of the atoms due to their hyperfine interactions and applied magnetic fields; (b) spin relaxation in the ground state due to diffusion to the walls, S damping [9] and Carver-rate damping [10] in the buffer gas, and spin exchange between colliding alkali-metal atoms [9,11]; (c) excited-

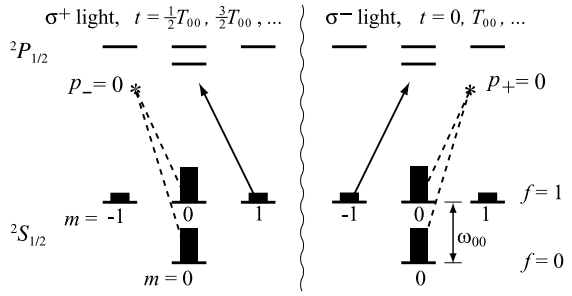

FIG. 1. Push-pull pumping of a hypothetical alkali-metal atom, with nuclear spin quantum number $I = 1/2$. In the left panel, atoms are hit by an LCP pulse at a time when the electron spin is $\langle S_z \rangle = -1/2$ for the superposition state $|\psi\rangle$ of Eq. (1), and for the end state $|fm\rangle = |1, -1\rangle$. Neither state can absorb photons from the pulse (dashed lines), but the photons are absorbed strongly by atoms in the other end state $|1, 1\rangle$ (solid line). In the right panel, the atoms are hit by an RCP pulse half a cycle later, at a time when the electron spin is $\langle S_z \rangle = 1/2$ for the superposition state $|\psi\rangle$ and for the end state $|1, 1\rangle$. Neither state can absorb photons from the pulse, but the photons are absorbed strongly by atoms in the other end state $|1, -1\rangle$.

state spin relaxation due to J damping by buffer gases [12,13]; (d) pressure broadening of the optical absorption line by buffer gases [14]; (e) the depopulation of the ground state due to absorption of light and its repopulation due to the spontaneous decay of excited atoms and quenching in molecular buffer gases like nitrogen [9]; (f) the modulation format of the light, its sideband structure, etc.

These phenomena can be modeled by considering their effects on the evolution of the density matrix of the atoms. For example, the evolution of the ground-state density matrix ρ is given by [9]

$$\frac{d\rho}{dt} = \frac{[H, \rho]}{i\hbar} + D\nabla^2\rho - \Gamma_{\rm sd}\left(\frac{3\rho}{4} - \mathbf{S}\cdot\rho\mathbf{S}\right) + \cdots. \quad (4)$$

The first three terms on the right-hand side of Eq. (4) denote evolution due to the ground-state Hamiltonian H, which includes hyperfine couplings and interactions with the applied magnetic field; diffusional relaxation with a diffusion coefficient D; and S damping at the rate $\Gamma_{\rm sd}$. The additional mechanisms listed above are denoted by $\cdots$. There are equations analogous to Eq. (4) for the density matrix of the excited state and for the optical coherence induced in the atoms by the pumping light.

Our modeling studies have led to a number of interesting predictions about push-pull pumping, many of which we have verified by experiment. The short pulses of the previous illustrative example are not needed at low buffer-gas pressures, as discussed in the next paragraph. There are only two essential requirements: (i) the time-averaged optical pumping rate must greatly exceed the ground-state relaxation rates and (ii) alternating polarization must be used.

For low gas pressures, where the hyperfine structure of the optical absorption lines is well resolved, it is sufficient to use a simple electro-optic modulation of linearly polarized monochromatic laser light to produce two equal-intensity spectral sidebands with a frequency difference of ω_{00}. One sideband should be approximately resonant for pumping out of the lower hyperfine multiplet and the second should be approximately resonant for pumping out of the upper hyperfine multiplet. If the resulting "two-wave" light is separated into right- and left-circularly polarized sub-beams, and if one sub-beam is delayed by $T_{00}/2$ with respect to the other, the combined beam can pump nearly all of the atoms into the 0-0 superposition state $|\psi\rangle$ of Eq. (1). In the language commonly used in CPT research, this is equivalent to a combination of two Λ-pumping schemes, one with σ^+ (RCP) and the other with σ^- (LCP) light, where a 180° modulation phase difference is maintained between the two.

For higher gas pressures, where the hyperfine structure of the atomic absorption line is not resolved, commonly used frequency modulated (FM) light cannot excite CPT resonances [15,16], and it is necessary for the light to be amplitude modulated (AM). In this case, even for buffer-gas pressures of many atmospheres, push-pull pumping with pulsed light can pump almost all of the atoms into the 0-0 superposition state $|\psi\rangle$ of Eq. (1).

The "photon cost" n, which we define as the mean number of photons per atom that must be absorbed to produce the superposition state $|\psi\rangle$ of Eq. (1), can be readily calculated. It depends on the nuclear spin quantum

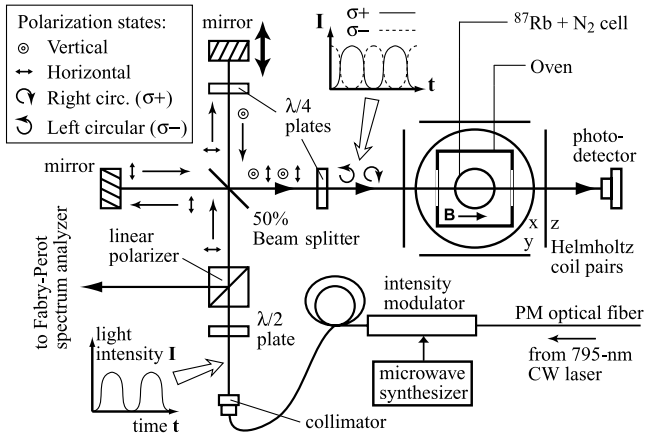

FIG. 2. Apparatus used to test push-pull pumping. A ^{87}Rb cell was placed into a temperature-stabilized oven. Three sets of Helmholtz coils produced a field B of about 2 Gauss in the direction of the probing beam. A monochromatic laser beam was amplitude-modulated (AM) at the 0-0 resonance frequency $\nu_{00} = \omega_{00}/2\pi = 6.84$ GHz by driving a Mach-Zehnder intensity modulator with microwaves at the frequency $\nu_{00}/2$. The sideband structure of the modulated light was monitored with a Fabry-Perot spectrum analyzer. The AM light was converted to light with alternating circular polarization using a Michelson interferometer (shown to the left of the oven).

number I of the atom and on the amount of polarization that is retained before the optically excited atoms decay. For example, for a nonquenching gas like He at pressures on the order of an atmosphere, most of the spin polarization of optically excited atoms is destroyed before spontaneous decay [12]. In the limiting case of complete destruction of excited-state spin polarization, we find $n = 4I + 1$. For a molecular buffer gas (like nitrogen) at pressures on the order of an atmosphere, quenching collisions return most of the nuclear polarization of the excited state back to the ground state, but almost none of the electron spin polarization is returned [9]. In this case the photon costs turn out to be $n = 3$ (exact), 9.67, 19.4, and 32.1 for the nuclear spin quantum numbers $I = \frac{1}{2}, \frac{3}{2}, \frac{5}{2}$, and $\frac{7}{2}$. In an atomic beam, the collisions are rare, but the excited-state hyperfine interactions are strong enough that the hyperfine coherences average to zero before the atom spontaneously decays. In this case, we find $n = 3.5$ (exact), 7.85, 13.3, and 19.7 [17].

We used the simple apparatus shown in Fig. 2 to experiment with push-pull pumping. An external-cavity, single-mode diode laser (Toptica DL100) was tuned to the 795-nm $D1$ line of Rb. The linearly polarized laser light was coupled into a single-mode, polarization-maintaining (PM) fiber for intensity modulation in a commercial $LiNbO_3$ Mach-Zehnder modulator (EOspace, Model AZ 0K1-12-PFU-SFU-800). The modulator works by splitting the input beam and sending each sub-beam through one of two parallel electro-optic crystals, arranged to produce an optical phase retardation $\phi = \phi_0 + \phi_1 \sin\omega_m t$ in one sub-beam and an equal and opposite phase retardation in the other sub-beam. The phase shifts are controlled by a time-independent (or slowly varying) bias voltage V_0 (not shown), and by microwaves of power P_m and frequency $\nu_m = (2\pi)^{-1}\omega_m$ (chosen so that $\nu_m \approx \nu_{00}/2$). The bias phase is $\phi_0 \propto V_0$ and the modulation index is $\phi_1 \propto \sqrt{P_m}$. The sub-beams are recombined and made to interfere to produce amplitude-modulated light, which exits the modulator via a single-mode optical fiber. The sideband structure was monitored with a commercial confocal Fabry-Perot spectrum analyzer (Coherent, Model 33-6339, 30 GHz free spectral range).

The amplitude-modulated light of fixed linear polarization was converted to light of alternating polarization with a Michelson interferometer. The sub-beam in one arm of the interferometer passed twice through a $\lambda/4$ phase retardation plate, which converted the linear polarization from horizontal to vertical. For ^{87}Rb, the 0-0 resonance frequency is $\nu_{00} = 6.84$ GHz and the microwave wavelength is $\lambda_{00} = 4.39$ cm, so a displacement of one of the mirrors by $\lambda_{00}/4 = 1.1$ cm shifted the intensity peaks of one beam by half a hyperfine period with respect to the peaks of the other. The output beams, now in orthogonal states of linear polarization, were combined and passed through a second $\lambda/4$ plate, introduced to guarantee that the exiting beam of light is alternating between the states of right and left circular polarization. We used this exiting beam as a source of light for push-pull pumping of ^{87}Rb vapor in a glass cell.

Figure 3 shows the very large signal enhancement of the 0-0 CPT resonance produced by push-pull pumping in comparison to the CPT resonance induced by intensity modulated light of fixed circular polarization. The signals were obtained from a cell containing isotopically enriched ^{87}Rb and nitrogen gas at a pressure of 730 torr (defined at room temperature). Unlike the case of CPT with FM light, which has been most often used in the past and where the pumping efficiency diminishes rapidly for buffer-gas pressures in excess of a few hundred torr [15,16], the CPT signals with AM light are nearly independent of gas pressure. The conventional CPT signal of Fig. 3 is very small mostly due to the optical pumping of atoms into the end state $|aa\rangle$, or $|a, -a\rangle$, depending on the sign of the fixed circular polarization. Push-pull pumping eliminates this cause of signal degradation.

More systematic studies of push-pull and conventional CPT resonances are summarized by the experimental points in Fig. 4. The measurements were done with Rb cells containing much lower gas pressures than the cell of Fig. 3. Modeling calculations of the expected CPT signals, shown by the continuous lines in Fig. 4, are in good

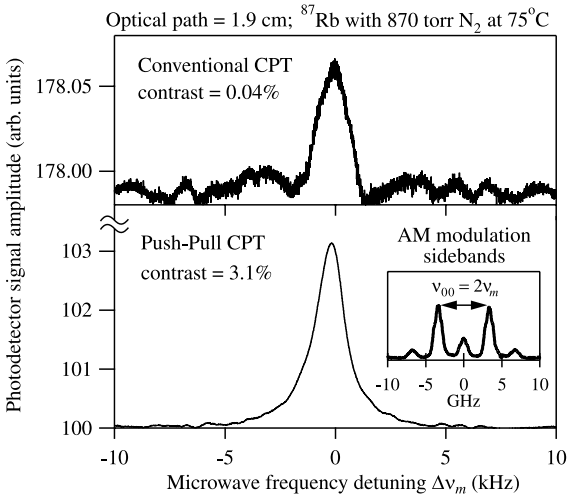

FIG. 3. Enhancement of the 0-0 CPT resonance with push-pull pumping. The top panel shows the conventional CPT signal (the intensity of the light recorded by the photodetector of Fig. 2) for amplitude-modulated light of fixed circular polarization. The CPT contrast obtained with push-pull pumping in the bottom panel is larger by a factor of about 77. The off-resonant signal amplitude is larger for the conventional signal (top panel) because a large fraction of the atoms are pumped into an end state [19]. Push-pull pumping produces no off-resonant polarization so the off-resonant transmission of the vapor is lower. The "contrast" is the increase of the resonant photodetector signal divided by the off-resonance signal. The inset shows the sideband spectrum of the amplitude-modulated light exiting the intensity modulator of Fig. 2.

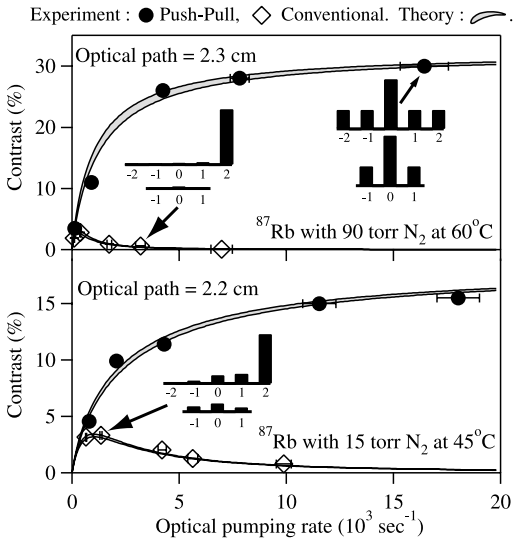

FIG. 4. CPT signal contrasts versus the pumping rate for push-pull and conventional pumping in low-pressure cells. The uncertainty of the theoretical curves, represented by the shaded area, is due to the uncertainty of the relaxation parameters. The insets show the modeled population distributions.

agreement with the experiments. Typical population distributions from the modeling are shown as insets. Conventional CPT signals first grow with increasing light intensity, as faster pumping overcomes relaxation processes. After reaching a peak, the signals decrease as the light pumps more and more atoms into one of the end states, the dark states for the light of fixed circular polarization [18]. Faster push-pull pumping, however, always increases the concentration of atoms in the superposition state $|\psi\rangle$ of Eq. (1), the only dark state for the light of alternating circular polarization. As shown in the top panel of Fig. 4, a 30% contrast was measured for the push-pull CPT resonance, a substantially larger figure than has ever been previously reported.

Because of birefringence in the glass windows of our oven and the glass walls of the vapor cell, the circular polarization was degraded and the peak spin of absorbed photons was measured to be $|s_z| = 0.87$. Taking this into account, our modeling calculations predict that 51% of the atoms were concentrated in the superposition state $|\psi\rangle$ of Eq. (1), for the top-right experimental point in Fig. 4. The predicted concentration for fast pumping and for peak $|s_z| = 1$ at these conditions is 100%.

In summary, we have demonstrated that push-pull pumping can be readily implemented in practice, and it produces much larger CPT signals than conventional methods. We have already achieved a ~50% concentration of the pumped ^{87}Rb atoms in the pure state of Eq. (1), with no further attempts to optimize the apparatus. Since our experimental observations are in good agreement with the modeling calculations, one can be confident in the prediction that push-pull pumping with perfect circular polarization can produce atomic ensembles close to the pure-state limit, for which $\rho^2 = \rho$. Using similar methods, one should also be able to concentrate atomic populations in other sublevels. Push-pull pumping should be useful in the applications, such as atomic clocks and quantum computing, that benefit from having the largest possible fraction of atoms in a well-defined quantum superposition state.

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