PHYSICAL REVIEW A VOLUME 28, NUMBER 2 AUGUST 1983

Polarization of an atomic sodium beam by laser optical pumping

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(Received 31 March 1983)

A highly polarized ^{23}Na atomic beam is produced by optical pumping with the use of either a multimode cw dye laser or a single-mode ring dye laser followed by a double-passed acousto-optic modulator. Electron-spin polarizations of 0.72 to 0.90 are obtained.

The use of laser optical pumping to produce polarized alkali atom beams is of current interest. Polarized atom beams are useful in atomic physics for collision experiments and are of interest in nuclear physics for polarized targets and ion sources. Optical pumping produces polarized atoms by the repeated absorption of circularly polarized light followed by spontaneous emission. The process results in the transfer of atoms to states with high M_F for absorption of σ^+ light where M_F is the z component of the total angular momentum F. Effective optical pumping requires that the product of light intensity times the atom-light interaction time is large enough that many cycles of absorption followed by spontaneous emission occur. In order to achieve maximum polarization, it is necessary either to have absorption from both ground hyperfine levels or to have absorption from one hyperfine level and to eliminate atoms in the other hyperfine level. Several experiments on the laser optical pumping of alkali beams have been previously reported. Baum, Caldwell, and Schroder[1] have polarized a ^6Li beam using a single-frequency dye laser with an acousto-optic modulator (AOM) to produce two light beams at two different frequencies, to pump both ground hyperfine levels. Both Hils, Jitschin, and Kleinpoppen[2] and Dreves *et al.*[3] have polarized a Na beam using a single-frequency dye laser to pump atoms in the upper hyperfine level and a 6-pole magnet to remove atoms in the lower hyperfine level. Recently, Dreves *et al.*[4] have polarized a Na beam with a single-frequency dye laser plus rf transitions to couple the two ground hyperfine levels. Dreves *et al.*[4] have also demonstrated the transfer of atoms between different Zeeman hyperfine states using high-frequency adiabatic transitions, so that atoms in the $F=2$, $M_F=2$ state can be transferred to a different state. In this paper we report the production of a highly polarized beam of Na atoms both by optical pumping with a multimode dye laser and with a single-frequency dye laser followed by a double-passed AOM.

We first discuss the optical pumping of a Na beam with a multimode laser. A beam of Na atoms, produced by effusion through a stainless-steel capillary bundle from a stainless-steel oven heated to 650 K, passes through cooled collimator apertures. The atomic Na beam has a flux of 10^{16} atoms/sec sr. The angular divergence of the resulting atom beam is 1.5×10^{-2} rad. The Na beam is optically pumped by a multimode dye laser in a magnetic field of 1 mT. The optical pumping region is 25 cm from the oven. Our Spectra Physics multimode dye laser, using R6G dye, has a measured bandwidth of $1.5-2.0\times10^{10}$ Hz and a measured power of 1.1 W at the Na beam when pumped with 5 W of light from a Spectra Physics Ar^+ laser operating on all lines. The dye laser beam is circularly polarized and has a diameter of 0.3 cm. The light beam intersects the atom beam axis at normal incidence and is reflected so that it makes three passes through the atom beam. After optical pumping the atom beam passes through two polarization measurement regions. The first polarization measurement is made by use of the method of laser-induced fluorescence (LIF) in an intermediate field as described by Dreves *et al.*[5] The LIF uses a very low-intensity single-frequency dye laser, linearly polarized parallel to the 50-mT magnetic field of an electromagnet. The frequency of the probe laser is scanned across the $D_1(3\,^2S_{1/2}\rightarrow 3\,^2P_{1/2})$ absorption line of the atom beam. The nuclear spin of Na is $I=\frac{3}{2}$ so that there are eight Zeeman hyperfine states in the $3\,^2S_{1/2}$ level. They are labeled by $i=1-8$ corresponding to the low-field quantum numbers $F=1$, $M_F=-1$; 1,0; 1,1; 2 $-$2; 2 $-$1; 2,0; 2,1; and 2,2, respectively. The eight Zeeman hyperfine states in the $3\,^2P_{1/2}$ level are similarly labeled by $j=1-8$. Atoms in state i are excited by the weak intensity probe laser to an excited state j according to the $\Delta M=0$ transition rule. An analysis of the fluorescence signal S_{ij} accompanying spontaneous decay of state j yields the value of n_i, the occupation probability of state i. In the second polarization measurement the Na beam passes through a 6-pole magnet and the Na beam flux is measured with use of a tungsten hot wire detector. The 6-pole magnet has a length of 11.8 cm and a field strength of 450 mT at the pole tip radius of 0.15 cm. Atoms with $M_J=\frac{1}{2}$ at high values of magnetic field have a larger transmission probability through the 6-pole than atoms with $M_J=-\frac{1}{2}$. The relative transmission solid angles of the 6-pole magnet for the eight ground Zeeman hyperfine states of Na have been measured by the use of LIF. This was done in a separate experiment in which the positions of the 6-pole and LIF region were interchanged. The results of trajectory calculations for the transmission solid angles of the eight Na ground hyperfine states agree well with the measured values obtained by use of LIF. The atom beam flux is measured following the 6-pole magnet for σ^+ and σ^- optical pumping and without optical pumping. Values for the state occupation probabilities n_i and the electron-spin polarization P_e are calculated with use of the three flux measurements along with the measured ratios of the transmission solid angles. The calculation assumes all atoms are in one of the three states 3, 7, and 8 for σ^+ optical pumping and in states 1, 2, and 4 for σ^- optical pumping. The analysis is similar to that described in Refs. 1 and 6 except that the measured transmission solid angles are used.

Table I shows the results using our multimode laser for optical pumping on either the D_1 or D_2 $(3\,^2S_{1/2}\rightarrow 3\,^2P_{3/2})$ absorption lines when the laser makes three passes through the atom beam. Figure 1(a) shows the LIF measurement without optical pumping, and Fig. 1(b) shows the measure-

TABLE I. Occupation probabilities n_i and electron-spin polarization P_e as determined by use of LIF or the 6-pole magnet. The LIF value of P_e includes measured values of n_i for atoms in states 1,2,4,5,6 even though these are not shown in the table. The 6-pole values assume $n_i = 0$ for states other than 3,7,8 as described in Ref. 6. LIF values are not available in parts (3) and (4) due to lack of a second single-frequency dye laser.

	n_3	n_7	n_8	P_e
(1) Multipole laser No. 1				
(a) D_2, 3-pass LIF	0.08	0.10	0.75	0.75
D_2, 3-pass LIF	0.10	0.13	0.77	0.78
(b) D_1, 3-pass LIF	0.06	0.14	0.71	0.74
D_1, 3-pass 6-pole	0.12	0.20	0.68	0.72
(2) Effects of angle on D_2, 1-pass pumping				
(a) Normal incidence LIF	0.06	0.12	0.69	0.70
(b) $\theta = 6.0 \times 10^{-2}$ rad LIF	0.15	0.13	0.58	0.56
(3) Multimode laser No. 2				
(a) D_2, 2-pass 6-pole	0.04	0.08	0.88	0.90
(4) Single-frequency laser with double-passed AOM				
(a) D_1, 1-pass 6-pole	0.08	0.10	0.82	0.83

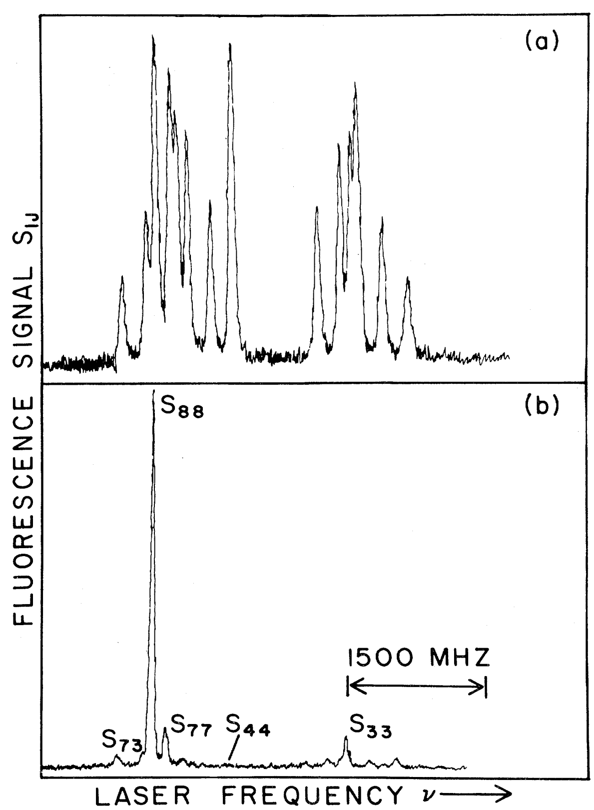

FIG. 1. (a) LIF measurement without optical pumping; (b) LIF measurement with σ^+ optical pumping [corresponding to entry (1)(a) in Table I].

ment with D_2 line optical pumping with three passes. We find that the occupation probabilities obtained by use of the 6-pole magnet are in good agreement with those obtained by use of the more accurate LIF for cases where the electron-spin polarization is 0.7 or greater. The results obtained by use of the 6-pole magnet do not agree as well with those obtained with LIF in experiments where $P_e < 0.7$ (not shown in Table I). Multiple-pass optical pumping both increases the Na polarization and the stability of the polarization. The polarization is maintained for several hours with only minor adjustments to the multimode laser. We have also studied single-pass optical pumping with a multimode dye laser as a function of the angle θ between the laser beam and the normal to the atomic beam axis in order to estimate the divergence of an atomic beam that can be optically pumped with a multimode dye laser. The results shown in Table I indicate that the polarization is reduced by about 20% when $\theta = 6 \times 10^{-2}$ rad.

The separation of the $F=1$ and 2 ground hyperfine levels is 1.772×10^9 Hz. The $3\,^2P_{1/2}$ level is split into two levels with $F'=1$ and 2 separated by 1.90×10^8 Hz. The $3\,^2P_{3/2}$ level is split into four hyperfine levels with $F'=3$, 2, 1, and 0 separated, respectively, by 5.60×10^7, 3.73×10^7, and 1.86×10^7 Hz. Thus for both the D_1 and the D_2 lines there are different absorption lines originating from each of the two ground hyperfine levels. Our Spectra Physics multimode dye laser has a longitudinal cavity mode separation of 4.0×10^8 Hz. A maximum of 50 cavity modes can oscillate within the output bandwidth of the laser. It is not necessary that all modes oscillate simultaneously. The value of the output frequencies can be changed by various adjustments, but the cavity mode separation is almost unchanged by these adjustments. The output frequencies of our laser cannot be adjusted so that the laser output simultaneously contains frequencies that are identical to line center absorption frequencies of any specific pair of absorption lines from the two ground hyperfine levels. The question arises therefore as to why the optical pumping of a Na atom beam is so successful. The photoabsorption cross section for a given transition is $\sigma(\nu) = \sigma_0 g(\nu - \nu_0)/g(0)$, where σ_0 is the cross section at line center ν_c, and $g(\nu - \nu_c)$ is the normalized absorption line shape. Feld *et al.*[7] have shown that the saturation intensity with optical pumping is approximately given by $I_s(\nu) = h\nu/\sigma(\nu)T$, where T is the atom-light interaction time. For a Lorentzian line shape with full width at half maximum of $\Delta\nu$,

$$\frac{\sigma(\nu)}{\sigma_0} = \left[1 + \frac{(\nu - \nu_c)^2}{(\Delta\nu/2)^2}\right]^{-1} ,$$

so that $I_s(\nu)$ increases as $|\nu - \nu_c|$ increases. For a Na beam, $\Delta\nu \simeq 1/2\pi\tau = 10^7$ Hz, where τ is the $3P$ radiative lifetime. The saturation intensity with optical pumping is smaller than in the absence of optical pumping by a factor of about $2\tau/T^7$. For our three pass measurements $2\tau/T$ is $\frac{1}{300}$. We estimate that $I_s(\nu_c)$, the saturation intensity with optical pumping on the D_2 line, is 0.03 mW/cm^2 at line center compared with a value of 9.6 mW/cm^2 without optical pumping. If our multimode laser has all 50 modes oscillating, a laser power at the Na atom beam of 1.1 W, and a beam diameter of 0.3 cm, then the average intensity of each mode is 300 mW/cm^2. Thus, for our intensity, and with optical pumping, saturation is expected to occur even far from line center. We believe this is why the multimode laser is effective in optically pumping the Na beam.

An improvement in the optical pumping with use of a multimode dye laser may be possible if the longitudinal cavity mode separation of the laser is reduced. This may increase the likelihood that Na absorption lines from both ground hyperfine levels are simultaneously saturated by a laser operating on many frequencies. The Coherent Radiation single-frequency dye laser used in the LIF measurement can be converted to standing-wave operation with a resultant longitudinal cavity mode separation of 2.0×10^8 Hz. The measured output bandwidth is 4.0×10^{10} Hz. The measured power at the Na beam is 1.3 W in a 0.5-cm-diam beam. The results of σ^+ optical pumping for both one and two passes with the use of the Coherent multimode dye laser are shown in Table I. Although LIF measurements of the occupation probabilities for the eight Zeeman hyperfine states were not made due to the lack of a second single-frequency dye laser, the values obtained through the 6-pole method indicate a higher polarization than was obtained with use of the Spectra Physics multimode laser.

We now discuss the second method for optical pumping of a Na beam, using a single-frequency dye laser followed by a double-passed AOM. Our Coherent Radiation single-frequency dye laser produces 600 mW of output power when pumped with 6 W from an Ar^+ laser operating on all lines. The ring laser is tuned to the D_1 absorption line of Na and the output beam is split into two beams which are incident in the first of two AOM's, AOM-1 (Harris Corp. H-191) drive at $\nu_1=6.30\times10^8$ Hz. The two beams incident on AOM-1 enter the crystal at angles θ and $-\theta$ from the normal to the crystal. The output of the modulator for each incident beam is an unshifted beam and a diffracted beam. The two diffracted beams are shifted by ν_1 and $-\nu_1$ from the laser output frequency ν_0. The angle θ is adjusted for a 60% diffraction efficiency for each incident beam. The process is repeated with the diffracted output beams from AOM-1 incident on AOM-2 (Intra Action AOM-228X) driven at $\nu_2=2.52\times10^8$ Hz.[8] The final output consists of two beams separated in frequency by $2(\nu_1+\nu_2)=1.772\times10^9$ Hz and with 80 mW in each beam. The two beams are circularly polarized and intersect in the optical pumping region. As the laser is tuned through the D_1 absorption line atoms in both ground hyperfine levels absorb laser radiation. Only one pass of the two light beams through the Na atom beam is used. The Na polarization is determined by use of the 6-pole magnet since a second single-mode laser is not available. The results of optical pumping with a single-frequency dye laser with the use of a double-passed AOM are shown in Table I. Rate equation calculations using our value of σ^+ light intensity and our interaction time of 3.0×10^{-6} sec predict that nearly all atoms will be pumped into the 2,2 state. This is strongly supported by the results in Table I.

In summary, a Na beam with high polarization has been obtained by two different methods of laser optical pumping. The use of multiple passes of a multimode dye laser through the Na beam results in good optical pumping. The use of a single-frequency dye laser followed by a double-passed AOM also results in good optical pumping.

We thank Dr. D. Mavis for his assistance with the 6-pole trajectory calculations. This work is supported in part by the Department of Energy.

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[8]Rather than use two AOM's, as we have done, one can use a single AOM with $\nu=8.86\times10^8$ Hz. We use two AOM's because we had previously purchased them for other experiments.