Repumping and spectroscopy of laser-cooled Sr atoms using the (5s5p)\(^3\)P\(_2\)–(5s4d)\(^3\)D\(_2\) transition

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Abstract
We describe repumping and spectroscopy of laser-cooled strontium (Sr) atoms using the (5s5p)\(^3\)P\(_2\)–(5s4d)\(^3\)D\(_2\) transition. Atom number in a magneto-optical trap is enhanced by driving this transition because Sr atoms that have decayed into the (5s5p)\(^3\)P\(_2\) dark state are repumped back into the (5s2)\(^1\)S\(_0\) ground state. Spectroscopy of \(^{84}\)Sr, \(^{86}\)Sr, \(^{87}\)Sr and \(^{88}\)Sr improves the value of the (5s5p)\(^3\)P\(_2\)–(5s4d)\(^3\)D\(_2\) transition frequency and determines the isotope shifts for the transition accurately enough to guide laser-cooling experiments with less abundant isotopes.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
Cold atom experiments require cycling transitions for efficient laser cooling and trapping. Depending on the level structure, atoms may be shelved into dark states during laser cooling, which removes them from the cooling cycle and can cause them to be lost from the trap. By applying laser light of the appropriate frequency to shelved atoms, it is possible to return these atoms to the cycling transition [1, 2]. This repumping process can increase atom number and density, which improves signal-to-noise ratios for most measurements, enables the study of collisional processes and is crucial for achieving quantum degeneracy [3–6].

In experiments with alkali-metal atoms, the dark states are ground-state hyperfine levels and repumping lasers can usually be generated with acousto-optic or electro-optic modulators from the laser used for cooling. In alkaline-earth-metal atoms such as strontium (Sr), atom population is trapped in highly excited metastable levels and independent lasers are necessary. Despite requiring additional lasers, alkaline-earth-metal atoms are interesting to study because they possess narrow optical transitions that can be used for optical frequency standards [7] and for attaining high phase-space density purely with laser cooling methods. In addition, they offer optical Feshbach resonances with relatively low loss that have the potential to provide a valuable tool for fine control of atomic interactions [7–9].

For Sr, the principal cycling transition for laser cooling operates between the (5s\(^2\))\(^1\)S\(_0\) and the (5s5p)\(^1\)P\(_1\) states (figure 1). Decay via the (5s5p)\(^1\)P\(_1\)–(5s4d)\(^1\)D\(_2\) transition [10] allows atoms to escape the cycling transition, and further decay from the (5s4d)\(^1\)D\(_2\) state results in atoms in the (5s5p)\(^3\)P\(_1\) and (5s5p)\(^3\)P\(_2\) states (henceforth \(^3\)P\(_j\)). \(^3\)P\(_1\) atoms return to the ground state and are recaptured in the MOT, but \(^3\)P\(_2\) atoms are shelved because of the 17 min lifetime of the \(^3\)P\(_2\) state [11].

Here, we describe a repumping scheme for Sr using the \(^3\)P\(_2\)–(5s4d)\(^3\)D\(_2\) transition at 3012 nm which has a historically difficult-to-reach wavelength in the mid-infrared (MIR). Lasers of this frequency based on optical parametric oscillators have recently become available due to advances in nonlinear optics and fibre lasers. A similar transition has been used to create a calcium MOT [12] operating on the 1978 nm \(^3\)P\(_2\)–\(^3\)D\(_3\) cycling transition. Other Sr repumping schemes are described in [13] or [14] that use a 707 nm laser to access the \(^3\)S\(_1\) state or in [15] which uses 496 nm light, requiring second harmonic generation, to excite to the (5s5d)\(^3\)D\(_2\) state. As difference frequency generation lasers are also now available for the 3 \(\mu\)m region, and as technology improvements promise to increase access to the MIR, the \(^3\)P\(_2\)–(5s4d)\(^3\)D\(_2\) transition may be a good solution for Sr repumping.
The OPO laser enables the repumping scheme outlined in the text by pumping atoms that have leaked from the \(^1\text{P}_1\) to the \(^3\text{D}_2\) state up to the \(^1\text{D}_2\) state, thus allowing decay to the \(^1\text{P}_1\) state and subsequent return to the \(^1\text{S}_0\) ground state. The main cycling transition operates on the \(^1\text{S}_0\) to \(^1\text{P}_1\) transition, and time-of-flight absorption imaging of ground-state atoms is performed using 461 nm light.

We also determine an improved value of the transition frequency and perform spectroscopy of the \(^3\text{P}_2\)\(^\rightarrow\)\(^5\text{S}_4\text{d}\) transition for \(^{84}\text{Sr}\), \(^{86}\text{Sr}\), \(^{87}\text{Sr}\) and \(^{88}\text{Sr}\). Using these spectra, we assign isotope shifts for the \(^{84}\text{Sr}\), \(^{86}\text{Sr}\) and \(^{87}\text{Sr}\) transition, relative to the \(^{88}\text{Sr}\) transition, with sufficient accuracy to inform laser cooling of the less abundant isotopes.

2. MOT loading using the repumping laser

Our experiment begins similarly to previously published work [13, 16, 17]. Without the repumping laser applied, as many as \(50 \times 10^6\) \(^{88}\text{Sr}\) atoms are trapped in a magneto-optical trap (MOT) operating on the 461 nm cycling transition between the \(^1\text{S}_0\) and \(^1\text{P}_1\) states. Time-of-flight absorption imaging is also performed using the \(^1\text{S}_0\)\(^\rightarrow\)\(^1\text{P}_1\) transition. Light at 461 nm is produced by frequency doubling via KNbO\(_3\) in a linear enhancement cavity [19]. The MOT beams, red-detuned by 60 MHz from resonance and with intensity-per-beam \(I = 2.3\ \text{mW cm}^{-2}\), yield atom samples with a temperature of about 2 mK, a density on the order of \(10^{10}\ \text{cm}^{-3}\), and a 1/\(e^2\) radius of about 1 mm. We also trap other Sr isotopes [18], \(^{85}\text{Sr}\) (<3 \(\times 10^5\) atoms), \(^{86}\text{Sr}\) (10 \(\times 10^5\) atoms) and \(^{87}\text{Sr}\) (5 \(\times 10^6\) atoms). These numbers are obtained with a 2D collimation stage of laser cooling immediately after the atom source and before the Zeeman slowing stage. Omitting the 2D collimation reduces the atom loading rate by about a factor of 6.

We produce 3 \(\mu\text{m}\) light for repumping and spectroscopy using a laser based on optical parametric oscillation (OPO) which is seeded by a fibre laser at 1.06 \(\mu\text{m}\) [20]. Our experiments only require a minimal amount of power, typically about 4 mW incident on the atoms, and the beam has a 1/\(e^2\) radius of about 3 mm. We frequency stabilize the laser to 0.002 \(\text{cm}^{-1}\) precision using a wavemeter that was calibrated against the ammonia spectrum.

As described earlier, the cycling transition used for the MOT is not closed because of leakage from the \(^1\text{P}_1\) state, leading to shelving of atoms in the \(^3\text{P}_2\) state. Figure 2 shows the number of atoms as a function of the MOT loading time with and without the repumping laser applied. We show the main isotope, \(^{88}\text{Sr}\), and also results from the least abundant stable isotope, \(^{84}\text{Sr}\), for which repumping is crucial for most experiments.

We analyse these data using the time-dependent number equation for MOT loading:

\[
N(t) = L_N - \Gamma N - \beta' N^2,
\]

Here, \(N\) is the number of atoms, \(L_N\) is the loading rate of atoms into the MOT, \(\Gamma\) is the one-body loss rate and \(\beta' = \beta/(2\sqrt{2}V)\), where \(\beta\) is the two-body loss constant and \(V = \int d^3r\ e^{-r^2/\sigma^2}\) is the effective volume for two-body processes (\(\sigma\) is the \(1/\sqrt{2}\) radius and \(r\) is the position). The solution to this differential equation is

\[
N(t) = N_{ss}(1 - e^{-\gamma t})/(1 + \chi e^{-\gamma t}),
\]

with \(\gamma = \Gamma + 2\beta' N_{ss}\), \(N_{ss}\) being the steady-state number of atoms and \(\chi\) the measure of the relative contributions of the one- and two-body loss coefficients:

\[
N_{ss} = -\frac{\Gamma + \sqrt{\Gamma^2 + 4\beta' L}}{2\beta'}
\]

and

\[
\chi = \frac{\beta' N_{ss}}{\beta' N_{ss} + \Gamma}.
\]

Using this model, we determine the fits shown in figure 2.

In the absence of the repumping laser, the loading curves are fit by the exponential limit of \(\beta = 0\), with \(\Gamma\) on the order of 10 s\(^{-1}\), consistent with optical pumping of atoms to the \(^1\text{P}_1\) state by the MOT laser [21]. In figure 2(A), the number of \(^{88}\text{Sr}\) atoms is kept artificially low, both with and without the repumping laser, by removing the 2D collimation beams. A full fit for \(^{88}\text{Sr}\) and \(^{84}\text{Sr}\) using equation (2) yields a value of \(\beta\) consistent with zero, indicating that the two-body losses are negligible at this low number. The repumping...
enhancements for both isotopes in this case are approximately equal to 20. The observed lifetimes (1/Γ) of 0.96 ± 0.01 s for 88Sr and 1.40 ± 0.06 s for 84Sr are significantly less than the 20 s lifetime we observe for 3P2 atoms in the magnetic trap formed by the MOT quadrupole magnets [13]. This small level of repumping may arise from the escape of atoms from the trapping region before atoms are returned to the ground state.

Figure 2(B) shows the loading curve for 88Sr with the 2D collimation beams aligned and an overall larger number of atoms. We suspect that several factors contribute to the decreased enhancement. A fit using equation (2) yields β = 6 ± 2 × 10−10 cm3 s−1 and Γ = 2.4 ± 0.1 s−1, indicating a contribution from two-body light assisted losses. The increased Γ also implies additional losses for larger atom number. Increased escape from the trap before repumping may result as implied by the fact that the optical depth for trapping light is on the order of unity, and the atom cloud increases significantly in size to ~2 mm and displays a non-Gaussian shape. The observed value of β is in agreement with [21], but no attempt was made to estimate systematic uncertainties arising from the distorted shape of the atoms’ density distribution.

3. 3P2–3D2 spectroscopy of laser-cooled Sr atoms

Using the repumping of atoms, we performed spectroscopy of the 3P2–3D2 transition for all the stable isotopes of Sr. For this study, we observe the repumping enhancement in the steady-state number of MOT atoms, although trapping of 3P2 atoms in the magnetic trap formed by the quadrupole magnets of the MOT [13] can affect the results. Scanning the laser across the resonance frequency of the repumping transition changes the number of atoms imaged (figure 3). The structure of the even isotopes, 88Sr, 86Sr and 84Sr, is simpler than that of the odd isotope, 87Sr, because the even isotopes have nuclear spin equal to zero.

At low repumping laser intensity, the spectra of 86Sr and 88Sr (see the inset of figure 3) reveal the structure arising from Zeeman splitting due to the 50 G cm−1 magnetic field gradient of the MOT magnetic coils. The detailed dynamics of the repumping process are beyond the scope of this paper. We suspect that at low repumping laser intensities used for these isotopes, the repumping is slow enough that atoms escape the region of the MOT unless they are in the mJ = 2 and mJ = 1 sub levels and are magnetically trapped [13], and mJ = 2 is more populated because it is trapped more strongly. The double peaks we observe are likely due to transitions from the mJ = 2 state of 3P2 to the mJ = 2 and mJ = 1 states in the 3D2 manifold. The observed splitting matches what one would expect from the known magnetic moments of the upper and lower levels, the magnetic field gradient, and the temperature of atoms in the MOT [13]. This simple model allows us to determine the position of the unperturbed resonances (figure 3, inset). For 84Sr and 87Sr, all the repumping laser power is necessary to achieve signal because of the low natural abundance of 84Sr (0.56%) and the poor repumping efficiency of 87Sr, and no structure is observed. For these isotopes, the unperturbed resonances are taken as the centre of the line.

The 87Sr spectrum shows the hyperfine structure because it has a nuclear spin of I = 9/2, but since the spectra are taken at high repumping laser intensity, no magnetic sub-levels are observed. We calculate the positions of the hyperfine states using the Casimir formula:

$$\Delta E_F = \frac{A}{2} + \frac{B}{2} \left[ \frac{3/4 K (K+1) - J(J+1) J(J+1)}{(2I-1)(2J-1)} \right].$$

with K = F(F + 1) − J(J + 1) − I(I + 1) and the values of the magnetic dipole and electric quadrupole factors (A and B, respectively) taken from [22] for the 3D2 level and from [23] for the 3P2 level. For this transition J = 2 and I = 9/2, and the total angular momentum, F, varies from 5/2 to 13/2 for both the upper and lower states of the transition. We overlay the calculated positions on the observed spectrum to assign the experimental peaks to the calculated positions.

To calibrate the wavemeter absolutely, we perform absorption spectroscopy of ammonia [24] in a gas cell at room temperature and ~1 Torr. Expected pressure shifts on the order of 1 MHz [25] are negligible. We correct for the systematic error in our wavemeter when stating our measurements of the Sr transition. We find the resonance wave number of the 3P2–3D2 transition in 86Sr to be 3320.226 ± 0.0025 cm−1, which is a small shift and improvement over the previously available
value of 3320.232 cm\(^{-1}\) [26]. Our uncertainty arises from statistical uncertainty in fitting the spectral features and from drifts in the wavemeter calibration.

Table 1 lists the isotope shifts relative to \(^{88}\text{Sr}\). The specification for the repumping laser linewidth on a short timescale (<10 ms) is less than 1 MHz, and the narrowest features we have studied with the laser experimentally are \(\sim 30\) MHz. The uncertainties reflect uncertainty in finding the unperturbed resonance positions from the spectra. Figure 4 compares our values for the isotope shifts to previous isotope shift measurements on the \(^{1}\text{S}_0\text{−3P}_1\) [27] and \(^{1}\text{S}_0\text{−3P}_1\) [28] \(\text{Sr}\) lines with a King plot [29, 30] of the modified isotope shift (\(\delta v_M\))

\[
\delta v_M = (\delta v_{\text{IS}} - \delta v_{\text{NMS}}) \times \frac{A_1 A_2}{A_1 - A_2}
\]

where \(A_1\) and \(A_2\) are the mass numbers in atomic mass units (amu) of the isotopes, \(\delta v_{\text{IS}}\) is the observed isotope shift, and \(\delta v_{\text{NMS}} = (v_{cm}/m_p) \times (A_1 - A_2)/A_1 A_2\) is the normal mass shift caused by the reduced mass of the atom \((v\) is the frequency of the transition; \(m_0\) and \(m_p\) are electron and proton masses). Within the error, this King plot shows the expected linear relations between the isotope shifts for the different transitions.

4. Conclusion

In conclusion, we have shown repumping of all stable isotopes of \(\text{Sr}\) using the \(^{3}\text{P}_2\text{−3D}_2\) transition. Additionally, we have measured the isotope shift of the \(^{3}\text{P}_2\text{−3D}_2\) transition for \(^{84}\text{Sr}\), \(^{86}\text{Sr}\) and \(^{88}\text{Sr}\) and provided an improved value for the \(^{3}\text{P}_2\text{−3D}_2\) transition wavelength of \(^{88}\text{Sr}\).

Acknowledgments

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