All-optical single-species cesium atomic comagnetometer with optical free induction decay detection

Yucheng Yang1 · Teng Wu1 · Jingbiao Chen1 · Xiang Peng1 · Hong Guo1

Received: 19 May 2020 / Accepted: 12 February 2021 / Published online: 25 February 2021
© The Author(s), under exclusive licence to Springer-Verlag GmbH, DE part of Springer Nature 2021

Abstract
Atomic comagnetometers, which measure the spin precession frequencies of overlapped species simultaneously, are widely applied to search for exotic spin-dependent interactions. Here we propose and implement an all-optical single-species Cs atomic comagnetometer based on the optical free induction decay (FID) signal of Cs atoms in hyperfine levels \( F_g = 3 \) and 4 within the same atomic ensemble. We experimentally show that systematic errors induced by magnetic field gradients and pump light are highly suppressed in the comagnetometer. With further optimization, the single-species comagnetometer has the potential to be applied in searches for exotic spin-dependent interactions.

1 Introduction

The spin-magnetic interaction is applied by atomic magnetometers to detect the magnetic field with a high sensitivity by measuring the atomic spin polarization precession frequency [1]. But to detect the non-magnetic spin-dependent interactions, the impact of magnetic field variation should be eliminated, and therefore the atomic comagnetometer scheme was presented [2–4]. By detecting the overlap of spin precession frequencies in the same magnetic environment, comagnetometers can suppress the impact of magnetic field variation in common mode, which may benefit the measurement of non-magnetic spin-dependent interactions in fundamental physics [5], such as tests of CPT and Lorentz invariance [6–8], measurement of permanent electric dipole moments (EDMs) [9–11], and searches for exotic spin-gravity interactions [12–16].

The magnetic field gradient is one of the main factors that lead to the systematic errors in comagnetometers. Due to the difference in polarization [17], gravity [18] and/or thermal diffusion rate [19], the species may have different average positions in the magnetic environment. If the magnetic field gradients exist, different species may sense different magnetic fields, and the magnetic field variations in common mode can not be suppressed. Therefore, comagnetometers with various species will suffer from the accuracy reduction from the magnetic field gradients.

Measures are taken to fix the errors induced by magnetic field gradients, such as monitoring the Larmor frequency shift as a function of the applied magnetic field gradients [20], or correcting for the magnetic field gradients via theoretical calculations [17]. But these numerical methods are dependent on other parameters, such as the size of the atomic vapor cells, and may bring calibration errors. The scheme adopting alkali metal atoms (\(^{85}\)Rb and \(^{87}\)Rb [12, 13]) can seemingly almost root out the influence from the magnetic field gradients, because the fast diffusion rates of gas atoms will lead to almost same average positions. But the frequency shift induced by the magnetic field gradients [21], which is related to the gyromagnetic ratio [22], can still degrade the performance of the \(^{85}\)Rb and \(^{87}\)Rb comagnetometer. To eliminate the systematic errors from magnetic field gradients, the single-species scheme was proposed in a nuclear-spin comagnetometer based on a liquid of identical molecules [14], and the comagnetometer was experimentally shown to have suppressed systematic errors from magnetic field gradients. The performance of the nuclear-spin comagnetometer is limited by the small polarization ratio of the nuclear spins.

Laser fields also produce systematic errors in comagnetometers. The spin-precession frequencies of atoms will...
be shifted by the light (the light shift effect [23]), which accounts for the shifted result of the measured magnetic field. Furthermore, atoms illuminated by the laser light will have larger relaxation rate because of power broadening, which will degrade the systematic sensitivity. Consequently, laser light may deteriorate the accuracy and sensitivity of each magnetometer, and the errors caused by pump light and probe light should be calibrated carefully [12, 13].

In this paper, we put forward and carry out the single-species comagnetometer scheme making use of free induction decay (FID) signal of atomic spin polarization in an all-optical [24] nonlinear magneto-optical rotation (NMOR) [25] Cs magnetometer. Compared with comagnetometers with different overlapped species [12, 13, 17, 19, 20], our single-species atomic comagnetometer can suppress the systematic errors induced by magnetic field gradients, considering the fast atomic diffusion rate and the almost identical gyromagnetic ratios of the two hyperfine levels \( F_g = 3 \) & 4. Compared with the nuclear-spin comagnetometer [14, 26], our atomic comagnetometer has higher spin polarization ratio, and is promising to have a better signal-to-noise ratio (SNR).

A similar \(^{87}\text{Rb}\) atomic comagnetometer has been implemented by our group [16]. As is shown in Table 1, Cs atoms are preferred for three reasons:

1. The difference in gyromagnetic ratio of hyperfine levels in the ground state \( \Delta \gamma \) is large, which means that, the required magnetic field to make the MR signals resolvable is small.

2. The splitting of hyperfine levels in the ground state \( \Delta \nu_{\text{HFS}} \) is the largest in alkali metal atoms commonly used for magnetometers, which corresponds to the lowest nonlinear Zeeman shifts of magnetic sublevels in the same applied magnetic field, according to the Breit–Rabi formula [27].

3. The vapor pressure \( P_v \), at room temperature (RT, 298 K) is the largest, which will result in the largest MR signal amplitudes and best systematic SNR in the same temperature.

Recently, we found that a dual frequency Cs spin maser of a similar scheme was presented [28], in which the authors experimentally show that the systematic errors from light field still exist. In our system, the influence of the pump light, such as light shift and cross talk between hyperfine Zeeman sublevels, is almost eradicated because the pump light is blocked when detecting the optical free induction decay (FID) signal.

### 2 Experimental scheme

The experimental setup is shown in Fig. 1a. At the center of our system is a self-made paraffin coated cylinder Cs vapor cell (diameter = 2.5 cm, length = 2.5 cm), which was manufactured by ourselves. The longitudinal spin relaxation time of the Cs vapor cell was measured to be \( T_1 \approx 3.3 \) s, which is limited by “uniform relaxation” [33], i.e., the exchange of alkali atoms between the volume and the stem. The anti-relaxation coatings in the inner surface can effectively reduce the relaxation due to wall collisions. The Cs vapor cell is located in a seven-layer magnetic shield (manufactured by Beijing Zero-Magnet Technology Co., Ltd) made of a 1-mm thick high-permeability alloy. The temperature of the shield is stabilized at 22 °C to provide the Cs vapor cell a stable temperature environment, yielding vapor density of \( n \approx 3.5 \times 10^{10} \) atoms/cm\(^3\). Within the shield is a set of three-dimensional Helmholtz coils, driven with a current source (Krohn-hite Model 523 calibrator, stability ±1 ppm within 24 hours) to generate a bias DC magnetic field.

In order to measure the spin precession frequency of Cs atoms in \( F_g = 3 \) & 4, there are two main processes in this comagnetometer – preparation and measurement of atomic spin polarization (see Fig. 1b).

During the preparation of atomic spin polarization (step 1 & 2 in Fig. 1b, duration = 1 s), Cs atoms are illuminated with a left-circularly polarized pump light propagating along \( -\hat{x} \) (orthogonal to \( \hat{z} \), which is along \( \hat{z} \)) tuned to the center of the Doppler-broadened Cs D1 \( F_g = 3 \rightarrow F_g = 3 \) resonance, where \( F_g = 3 \) is the ground-state hyperfine level and \( F_g = 3 \) is the excited-state hyperfine level (see the red arrow in Fig. 1c). The 895 nm D1 pump beam is generated with a distributed Bragg reflector laser diode (Photodigm PH-895-DBR-080-T8). The peak value of the pump light power is \( \approx 3.75 \) mW, and the beam size is \( \approx 4 \) mm\(^2\). In the pumping process, most atoms in \( F_g = 3 \) are depopulated by the pump light, except for atoms in Zeeman sublevel \( m = +3 \); while because the excited atoms will repopulate to all Zeeman sublevels except for \( m = -4 \) in \( F_g = 4 \), the 1-order polarizations (orientations) in both hyperfine levels \( F_g = 3 \) and 4 are generated. To polarize the spin in both hyperfine levels, the pump light has its amplitude modulated at Larmor frequency of \( F_g = 4 \) and 3 successively with an acoustooptic modulator (AOM, ISOMET M1250-T150L-0.5). The duty cycle of the modulation is chosen at 20% to maximize

---

**Table 1**: Some data of alkali metal atoms commonly used for magnetometers

| Isotopic | \( \Delta \gamma \) (mHz/nT) | \( \Delta \nu_{\text{HFS}} \) (GHz) | \( P_v \) (Torr, RT) |
|----------|-------------------------------|------------------------------------|---------------------|
| \(^{39}\text{K}\) [29]       | 3.973                         | 0.461                              | \( 10^{-7.6} \)     |
| \(^{85}\text{Rb}\) [30]      | 8.220                         | 3.035                              | \( 10^{-6.4} \)     |
| \(^{87}\text{Rb}\) [31]      | 27.857                        | 6.835                              | \( 10^{-6.4} \)     |
| \(^{133}\text{Cs}\) [32]     | 11.165                        | 9.192                              | \( 10^{-5.8} \)     |
the spin polarization with a relatively long transverse spin relaxation time $T_2$ [34]. Note that, to make the MR signal amplitudes of $F_g = 3$ and 4 comparable, the spin in $F_g = 4$ must be polarized in step 1, and later to polarize the spin in $F_g = 3$ in step 2; otherwise, because of the faster relaxation rate and lower polarization ratio for atoms in $F_g = 3$, the MR signal amplitude of $F_g = 3$ is about one order of magnitude smaller than that in $F_g = 4$.

During the measurement of atomic spin polarization (step 3 in Fig. 1b, duration = 2 s, longer than the spin polarization relaxation time, which is about 106 ms for $F_g = 4$ and 53 ms for $F_g = 3$ in our experiments), the pump beam is blocked with the AOM, and a linearly polarized probe beam propagating along $-\hat{y}$ travels through the Cs vapor cell and into a polarimeter. The 852 nm D2 probe beam is produced with a tunable external-cavity diode laser (New Focus TLB-6817) with an isolator (Thorlabs IO-3D-850-VLP, not shown in Fig. 1a). The spin precession frequencies of atoms in $F_g = 3 & 4$ are measured by observing optical rotation of the probe light. The probe light is 5 GHz blue-detuned from the center frequency of the Doppler-broadened Cs D2 $F_g = 3 \rightarrow F_e = 4$ resonance (see the blue arrow in Fig. 1c), the power is $\approx 1.6$ mW, and the beam size is $\approx 4$ mm$^2$. The probe beam is split with a Wollaston prism and detected with a balanced detector (Thorlabs PDB210A). The optical FID signal from the balanced detector is sampled with a balanced detector (Thorlabs PDB210A). The optical FID signal is transformed into MR signals in spectrum via Fourier transformation and later fitted in the processor computer (1 s, this step can be done offline).

After the data acquisition, the MR signals are acquired through the Fourier transformation of the FID signal. The data analysis (step 4 in Fig. 1b, duration = 1 s) is conducted by fitting the MR signals to an overall Lorentzian profile:

$$L(f) = \sqrt{\left[\sum \frac{A_F}{1 + \left(\frac{f - f_F}{\nu_F}\right)^2}\right]^2 + \left[\sum \frac{B_F(f - f_F)/\nu_F}{1 + \left(\frac{f - f_F}{\nu_F}\right)^2}\right]^2},$$  

(1)

where $L(f)$ is the theoretical form of the overlapped MR signals in spectrum, $A_F$ and $B_F$ represent the amplitudes of the imaginary and real part of the signals, respectively, $f_F$ is the corresponding Larmor frequency of corresponding hyperfine level $F$, and $\nu_F$ is the resonance width (half width at half-maximum, HWHM) of the MR signal. All steps take 4 seconds in total.
To measure the non-magnetic spin-dependent interactions, comagnetometers should operate under proper magnetic field $B_0$, which makes the two MR signals resolvable in the spectrum and meanwhile the broadening in the widths of MR signals due to nonlinear Zeeman effect is not obvious. Improper magnetic field may degrade the accuracy and/or sensitivity of each magnetometer in the comagnetometer system, thus deteriorate the potential to set more stringent constraints on exotic spin-dependent couplings.

When the $B_0$ is small, the two MR signals of hyperfine levels in the ground state are not resolvable in the frequency spectrum, increasing the fitting error. Bad fitting may account for the error of the measured magnetic field and therefore, the failure in suppressing the magnetic field variations. To judge the accuracy of the two magnetometers, we construct the index $R$, the deviation between the measured frequency ratio and the theoretical deviation between the measured frequency ratio and the theoretical value, as

$$R = \frac{f_3}{f_4} / \frac{g_3}{g_4} - 1, \quad \frac{g_3 - 9g_4}{g_4 + 7g_4} \approx 1.003191233,$$

(2)

where $f_{3,4}$ are the measured center frequencies of MR signals for corresponding hyperfine levels, $g_{3,4}$ are the gyromagnetic ratios, $g_{i,j}$ is the Landé factor of electron and nuclei, and $|\gamma_{3,4}|$ can be calculated from Ref. [35]. As is shown in Fig. 2a, when the applied magnetic field $B_0 < 2 \mu T$, $R$ is too large, the accuracy of each magnetometer is limited by the fitting error. And when $B_0 > 2 \mu T$, the comagnetometer performance is immune to the magnetic field variations.

When the $B_0$ is large, the widths of the two MR signals will be broadened due to nonlinear Zeeman effect, and the sensitivity of the comagnetometer to detect non-magnetic spin-dependent interactions will be worsened. The sensitivity of the comagnetometer for measuring non-magnetic spin-dependent interaction is determined by the sensitivity of the two magnetometers that constitute the comagnetometer, and the sensitivity of a magnetometer is

$$\delta B \propto \frac{v}{\gamma \cdot \text{S/N}},$$

(3)

where $\gamma$ is the gyromagnetic ratio of working atoms, $v$ is the MR width (HWHM), and S/N is the signal-to-noise ratio of the system. However, due to nonlinear Zeeman effect, the amplitude becomes reduced, and the width becomes broadened, when the magnetic field $B_0$ grows. In the system, the noise is relatively constant, and the sensitivity of the magnetometer, which is depicted with Eq. (3), is mainly determined by the ratio of MR signal width and signal amplitude $v/S$. The dependence of this ratio on the magnetic field strength is shown in Fig. 2b, and the inset figure indicates that when the applied magnetic field $B_0 < 5 \mu T$, both magnetometers of $F_g = 3 \& 4$ have good sensitivity.

Overall, to achieve the optimal accuracy and sensitivity, the Cs comagnetometer should work at a magnetic field $B_0$ ranging from 2 $\mu T$ to 5 $\mu T$.

### 3 Estimation for systematic errors

For the single-species comagnetometer system, the index $R$ is related to the ability to search for non-magnetic spin-dependent interactions. According to Eq. (2), the systematic errors in measuring frequencies will finally effect on the index $R$ as

$$R_{\text{err}} = \frac{1}{f_4} \sqrt{\left(\frac{g_3}{g_4}\right)^2 f_{3,\text{err}}^2 + f_{4,\text{err}}^2},$$

(4)

where $f_{3/4,\text{err}}$ are the corresponding systematic errors in measuring the Larmor frequencies of hyperfine levels.

![Fig. 2](image-url) **a** The dependence of the index $R$ (deviation of measured frequency ratio from theoretical value) on the applied magnetic field $B_0$. When $B_0 < 2 \mu T$, the two MR signals are so close to each other in the spectrum that the fitting error will limit the accuracy on frequency measurement, leading to the large $R$. For $B_0 > 2 \mu T$, $R$ fluctuates within $\pm 4 \times 10^{-7}$. **b** The dependence of the width-to-amplitude ratio $v/S$ on the applied magnetic field $B_0$. The parameter $v/S$ is proportional to the sensitivity of frequency measurement. The inset shows the rescaled results for $B_0 < 5 \mu T$, indicating that within this range the measurements for Larmor frequencies of both hyperfine levels have good sensitivity. In our system, the optimal range of the applied magnetic field $B_0$ should be from 2 to 5 $\mu T$.
The sources of systematic errors in the system can be divided into four parts—the magnetic field, the (pump and probe) light fields, the atomic collisions, and the Earth rotation. In this section, we focus on the estimations for systematic errors from the magnetic field and the light fields.

### 3.1 Magnetic field

We verified that our atomic comagnetometer system is immune to the magnetic field gradients, by measuring the index $R$, which is depicted with Eq. (2). Figure 2a also shows that, the systematic errors from the magnetic field gradients, which are common in traditional comagnetometers, are suppressed in our Cs comagnetometer. Assuming the difference in magnetic field experienced by atoms in $F_g = 3$ and 4 is $\Delta B$, the index $R$ is given by

$$R \approx \frac{\gamma_3(B + \Delta B)}{\gamma_3 B} \left( \frac{\gamma_3}{\gamma_4} - 1 \right) = \frac{\Delta B}{B}. \tag{5}$$

If the Cs comagnetometer is sensitive to the magnetic field gradients, $R$ should be proportional to $B^{-1}$. However, Fig. 2a shows that, at the level of the measurement uncertainty, the measured $R$ has almost no dependence on the strength of the bias field $B$.

The geometric phase effect [22], which is related to the magnetic field gradients, is highly suppressed in our system, too. The frequency shift due to the geometric phase effect is given by [22]

$$\Delta f_{F,geo} = \frac{(\gamma_f B)^2}{2} \left( \frac{\gamma_f B}{(\gamma_f B)^2 - f_r^2} \right), \tag{6}$$

where $B_r = \nabla B \cdot r/2$, $f_r = \bar{v}/2\pi r$, $\nabla B$ is the magnetic field gradients, $\bar{v}$ is the mean thermal velocity of atoms, and $r$ is the radius of the cell. In our system, the applied magnetic field is $B = 3463.8$ nT, the cell radius is $r = 1.25$ cm, the mean thermal velocity of Cs atoms at $22^\circ C$ is $\bar{v} = 216.7$ m/s, and the magnetic field gradient along the direction of $B$ is $\nabla B_r = 1.5$ nT/cm. Substitute these parameters into Eq. (6), and the frequency shifts due to the geometric phase effect are

$$\Delta f_{3,geo} = -4.59 \times 10^{-4} \text{ Hz},$$
$$\Delta f_{4,geo} = -4.52 \times 10^{-4} \text{ Hz}. \tag{7}$$

Combining Eqs. (4) and (7), the upper limit of the systematic errors induced by the geometric phase effect in the index $R$ is calculated to be

$$R_{err,geo} = 5.325 \times 10^{-8}. \tag{8}$$

The nonlinear Zeeman effect is another way for the magnetic field to cause systematic errors in the comagnetometer system. The energy shift of each Zeeman sublevels $|F, m_F\rangle$ for Cs atoms at the magnetic field $B$ is given by the Breit-Rabi formula [27] as

$$\Delta E(F, m_F) = g_i \mu_B m_F B \pm \frac{\Delta E_{HFS}}{2} \left( 1 + \frac{m_F x^2}{2} \right)^{1/2}, \tag{9}$$

where $\mu_B$ is the Bohr magneton, $\Delta E_{HFS} = h \cdot \Delta \nu_{HFS}$ is the energy splitting between ground-state hyperfine levels, $\pm$ corresponds to the magnetic field $F' = I \pm j$. The spin precession frequency of atoms in Zeeman sublevel $|F, m_F\rangle$ is given by

$$f(F, m_F) = \frac{\Delta E(F, m_F) - \Delta E(F, m_F = 1)}{h}, \tag{10}$$

and the MR signal of hyperfine level $F$ can be treated as the overlap of $2F$ MR signals with different frequency. In our system, because the pump light is left-circularly polarized and tuned to the center of $F_g = 3 \rightarrow F_e = 3$ transition, atoms in $|F_g = 3, m_F = -3 \sim 2\rangle$ will be depopulated to $|F_e = 3, m_F = -2 \sim 3\rangle$. The excited atoms in $|F_e = 3, m_F = -2 \sim 3\rangle$ will spontaneously transit back to the Zeeman sublevels $|F_g = 3, m_F\rangle$ and $|F_g = 4, m_F = -3 \sim 4\rangle$ in the ground state. Atoms repopulating in $F_g = 3$ will be depopulated again. In the steady state, atoms in $F_g = 3$ mostly populate in $m_F = 3$, and the MR frequency is given by

$$f_{3,NLZ} = \frac{\Delta E(3, 3) - \Delta E(3, 2)}{h}. \tag{11}$$

The situation in $F_g = 4$ is more complicated. Due to the same transition rate for repopulation $|F_e = 3, \pm m_F\rangle \rightarrow |F_g = 4\rangle$, the contribution of atoms repopulating in $F_g = 4$ from $|F_g = 3, m_F = -2 \sim 2\rangle$ to overall MR signal is “neutralized”. The max frequency shift for $F_g = 4$ due to nonlinear Zeeman effect is given in the approximation that, only atoms repopulating in $F_g = 4$ via the transition $|F_e = 3, m_F = 3\rangle \rightarrow |F_g = 4, m_F = 2, 3, 4\rangle$ contribute to the comprehensive MR signal of $F_g = 4$, as

$$f_{4,NLZ} = \frac{\Delta E(4, 4) - \Delta E(4, 1)}{3h}. \tag{12}$$

Substituting the applied magnetic field is $B_0 = 3463.8$ nT in our system into Eq. 2, Eqs. (11) and (12), the upper limit of the systematic errors induced by nonlinear Zeeman effect is calculated to be

$$R_{err,NLZ} = 2.13 \times 10^{-8}. \tag{13}$$

The result is one order in magnitude larger than that in Ref. [16]. This may be because that the alignment is measured
in Ref. [16], and the nonlinear Zeeman effect is better suppressed in the comprehensive MR signal by averaging the MR signals of atoms in \(|F_g, \pm m_F|\).

### 3.2 Pump light

Our FID comagnetometer is proved capable of highly suppressing power broadening and light shift effect from the pump light, compared with single-species comagnetometers which obtain the MR signals by scanning the modulation frequency or magnetic field [16, 28]. The comparison is conducted by scanning the pump light power in FID mode and modulation-frequency-scan (MFS) mode at the same magnetic field \(B_0 = 3463.8 \text{ nT}\), using the same equipment. The information of the signal from polarimeter in MFS mode is acquired with a lock-in-amplifier (Stanford Research SR850) and later collected by a LabVIEW routine.

The atomic spin precession frequency, in the MFS mode, varies with the pump light power, because of the light shift effect in both \(F_g = 3\) (shown in Fig. 3a) and \(F_g = 4\) (shown in Fig. 3b). Within the range of the pump light power, the atomic spin precession frequency varies within a range of 6 Hz in \(F_g = 3\), and 0.4 Hz in \(F_g = 4\). The difference may come from the light configuration shown in Fig. 1c, which implies that the vector light shift is more obvious in \(F_g = 3\) due to the pump light. While in the FID mode, the atomic precession frequencies of both hyperfine levels fluctuate within 0.01 Hz when the pump light power is more than 0.2 mW.

As for the MR width (HWHM), in the MFS mode, it grows with the pump light power due to the power broadening effect in \(F_g = 3\) (shown in Fig. 3c) and \(F_g = 4\) (shown in Fig. 3d). With the increase of the pump light power, the MR width of \(F_g = 3\) grow steeply (31 Hz broadened at max pump light power), because the pump light interacts with atoms in \(F_g = 3\) directly; and the MR width of \(F_g = 4\) also increases slightly (1 Hz broadened at max pump light power), due to the Doppler broadening effect and the natural width of atoms. While in FID mode, the MR widths of both hyperfine levels are almost uniform (2.993 Hz for \(F_g = 3\), pink line; and 1.503 Hz for \(F_g = 4\), green line). The width difference mainly comes from the inconsistent relaxation rate induced by spin-exchange effect [36]. The broadening in MR signal width \(\Delta v_{\pm}\) due to spin-exchange collisions can be calculated as:

![Fig. 3](image-url)
where $\Gamma_{\pm}$ is the relaxation rate induced by spin exchange collisions, corresponds to the broadening in MR signals of hyperfine levels $F_g = I \pm 1/2$, $I$ is the nuclear spin (for Cs, $I = 7/2$), $T_{SE}$ is the spin-exchange time calculated by $T_{SE} = (n_{SE} \bar{v})^{-1}$, $n_{SE}$ is the cross section of spin-exchange collisions between Cs atoms, and $\bar{v}$ is the average thermal velocity of Cs atoms. In our experiments, the cell temperature is stabilized at $T = 22$ °C, and the broadening due to spin-exchange collisions are:

$$\Delta v_3 \approx 1.73 \text{ Hz}, \quad \Delta v_4 \approx 0.26 \text{ Hz},$$

the difference is 1.47 Hz in theory, which coincides well with the measured ~ 1.33 Hz difference in the width of the two MR signals in FID mode.

Due to the power broadening and light shift effect, as is underlined in Fig. 4a, the index $R$ in the MFS mode can be 200 times larger than that in the FID mode (4000:20). In the inset graph, the fluctuation of the index $R$ of FID mode, when the pump light power is less than 0.2 mW, may be due to the inadequate pumping. When the pump light power is low, the pumping rate is comparable to the atomic relaxation rate, and the atomic polarization of each hyperfine level is deficient, which refers to substandard SNR for fitting in data analysis. When the pump light power is large, the index $R$ fluctuates within $\pm 4 \times 10^{-7}$.

It is important to note that, even the AOM is not driven, some 895 nm light still arrives to the cell. The pumping effect of the residual 895 nm light can bring systematic errors by creating an imbalance of atomic population among the different Zeeman sublevels, which has been discussed in Sect. 3.1. The systematic errors should be less than what is estimated there, because the power of the residual 895 nm light is much less than that when the AOM is driven. When the peak value of the 895 nm light power is 3.75 mW with the AOM driven, the power of the 895 nm light is measured to be $\sim 0.95 \mu W$ when the AOM is not driven. The corresponding light intensity $I_{1st,0}$ is given by

$$I_{1st,0} = (0.95 \mu W)/(4 \text{ mm}^2) \approx 0.024 \text{ mW/cm}^2,$$

and the vector light shift effect it may bring is given by [37]

$$\Delta f_{\text{VLS}} = -\frac{|E_0^2|}{2} \sqrt{\frac{6F(2F + 1)}{(F + 1)} \epsilon \sin \phi} \times \frac{m_e}{2F} \{6S_{1/2}|\gamma E_{0}|(6P_{1/2})|^2 f(v),$$

where $E_0$ is the average optical electric field experienced by atoms, related to the light intensity as $\langle |E_0^2| \rangle = 2I_0/e\epsilon_0$, where $I_0$ is the light intensity and $\epsilon_0$ is the permittivity of

\[ \Delta v_{\pm} = \frac{\Gamma_{\pm}}{2\pi} \approx 1 \left( \frac{K \mp 1}{T_{SE}} \right) \left( 1 \mp K^2 - 1 \right), \quad K = \frac{I^2 + 2}{3I}, \]
vacuum, \(\epsilon = 1\) is the ellipticity of the left circularly polarized pump light, \(\phi\) is the deviation of the pump light direction from orthogonality to the magnetic field (in our system \(\phi \lesssim 1 \times 10^{-4}\) rad), \(\langle 6S_{1/2}\rangle_{F} = 3.17 e_{0}\) is the transition dipole matrix element between the \(6S_{1/2}\) and \(6P_{1/2}\) states, and \(\mathcal{F}(v)\) is sum of the imaginary part of three Voigt profiles [38, 39] with dimension \(\text{Hz}^{-1}\) as

\[
\mathcal{F}(v) = \sum_{J_{c}} \left\{ \begin{array}{ccc} 1 & 1 & 1 \\ F & F & F_{c} \end{array} \right\} \left\{ \begin{array}{ccc} J & J_{c} & 1 \\ F_{c} & F & F \end{array} \right\} \left(2F_{e} + 1\right) \Delta \Theta(\mathcal{V}(v)),
\]

where \(F_{e}\) represents the hyperfine levels in \(6P_{3/2}\), \(J_{c} = 1/2\) for D1 transition, \(I = 7/2\) for Cs and \(\mathcal{V}(v)\) is the Voigt profile corresponding to the interactions between atoms and light with different frequency \(v\). Taking the fluctuation of pump light frequency (~20 MHz within one day) into consideration, the vector light shift for MR signals frequencies are

\[
\Delta f_{\text{LS,pump, VLS}} \approx -2.61 \times 10^{-3} \text{ Hz},
\]

\[
\Delta f_{\text{LS,pump, VLS}} < 10^{-6} \text{ Hz},
\]

The frequency shift for \(F_{g} = 4\) is negligible because the pump light is ~9.2 GHz detuned from any \(F_{g} = 4 \rightarrow F_{e}\) transition. The corresponding upper limit of systematic errors can be calculated by Eq. (4) as

\[
R_{\text{err,pump, VLS}} \lesssim 2.16 \times 10^{-7}.
\]

The systematic errors due to vector light shift from the pump light can be further suppressed by stabilizing the pump light frequency.

Improper pump light modulation frequency is another reason for prominent frequency shift in MR signals [40]. This effect comes from the asynchronous optical pumping. If the pump modulation frequency is detuned from the spin precession frequency, the spin polarization will be tipped along the applied magnetic field and precesses around the fictitious magnetic field of the vector light shift induced by the pump light [13]. The fictitious magnetic field due to vector light shift is given by

\[
B_{F,\text{LS}} = \frac{\Delta f_{F,\text{VLS}}}{f_{F}}.
\]

The fictitious magnetic field generated by the pump light in Step 1 of Fig. 1b are calculated to be

\[
B_{\text{LS},1} \approx 2.94 \text{ nT}, \quad B_{\text{LS},2} \approx 0.
\]

The fictitious magnetic field modulated at frequency \(f_{\text{mod}}\) can be treated as an magnetic field rotating at frequency \(f_{\text{mod}}\) transverse to the real magnetic field generated by the coils. If the modulation frequency does not match the spin precession frequency, the phase of the spin precession signal will be shifted at \(\delta \varphi\). And the frequency shift \(\Delta f_{\text{asy}}\) due to asynchronous pumping is given by the phase shift of the spin precession signal over pumping time, taking the form as \([40]\)

\[
\Delta f_{\text{asy}} = \frac{\delta \varphi}{\tau} = \frac{1}{\tau} \frac{P_{y}}{P_{c}} = \frac{1}{\tau} \frac{\delta f \cdot f_{\text{LS}}}{(\delta f)^{2} + (\nu_{F})^{2}},
\]

where \(P_{y}, P_{c}\) is the polarization along axis of the rotating frame, with the expressions already given in Ref. [1], \(\delta f\) is the difference between the modulation frequency and the spin precession frequency, \(f_{\text{LS}}\) is the vector light shift from the pump light, \(\tau\) is the total time of the pump light, and \(\nu_{F}\) is the MR width (HWHM) of the hyperfine level. In our system, the time consumed to polarize atoms in one hyperfine level is 0.5 s, and the duty cycle of the modulation is 20%, so the total time of the pump light is \(\tau = 0.1 \text{ s}\). Because the vector light shift in \(F_{g} = 4\) is negligible, only \(F_{g} = 3\) will be affected by the asynchronous pumping. As is shown in Fig. 3c, the MR width of \(F_{g} = 3\) when the pump light is 0.75 mW (on average after AOQ) is \(\nu_{3} = 33 \text{ Hz}\). To minimize the systematic errors from asynchronous optical pumping, in our system, the pump light modulation frequencies are tuned to within \(\delta f \lesssim 2 \text{ Hz}\) of \(\omega_{4}\) and \(\omega_{3}\) during Step 1 & 2 in Fig. 1b. Substituting the parameters in our system into Eq. (23), the frequency shift for \(F_{g} = 3\) is calculated to be:

\[
\Delta f_{\text{asy}} = \pm 1.892 \times 10^{-4} \text{ Hz}.
\]

and the upper limit of the overall systematic errors of \(R\) from asynchronous optical pumping is

\[
R_{\text{err,asy}} = 1.564 \times 10^{-8}.
\]

**3.3 Probe light**

The systematic errors from the probe light are insignificant in the Cs comagnetometer. In general, the probe light may induce light shift and power broadening. Besides, the alignments generated by a linearly polarized light may be converted to orientations by external interactions, such as the magnetic field gradients, anisotropic collisions or electric fields [41].

The light shift effect induced by the probe light, can be analyzed in two parts: the vector element and the tensor element. The vector light shift is proportional to the product of the ellipticity \(\epsilon\) and the deviation for propagation direction the probe light from orthogonality to the magnetic field \(\theta\) [12]. In our system, the deviation is measured to be \(\theta \lesssim 1 \times 10^{-4}\) rad, and the ellipticity \(\epsilon\) of the linearly polarized probe light can be deduced from the nominal extinction ratio (100000:1) of the polarizer (Thorlabs, LPNR050-MP2) used after the 852 nm Laser and isolator. In our
experiments, the isolator and polarizer used after the 852 nm Laser are adjusted carefully to guarantee the linearity of the probe light. Assuming the polarizer is designed to transmit the horizontal polarized light, and the power ratio of the horizontal and vertical polarized light is no less than
\[
\frac{\langle |E_H|^2 \rangle}{\langle |E_V|^2 \rangle} \geq 10^6.
\]

(26)

For the linearly-polarized light, its left- and right-circularly polarized components, which can be depicted as
\[
E_L = \frac{E_H + i \cdot E_V}{\sqrt{2}} , \quad E_R = \frac{E_H - i \cdot E_V}{\sqrt{2}} ,
\]

(27)

the ellipticity \( \varepsilon \) refers to the normalized power imbalance between left- and right-circularly polarized light as
\[
e = \frac{\langle |E_L|^2 \rangle - \langle |E_R|^2 \rangle}{\langle |E_H|^2 \rangle + \langle |E_V|^2 \rangle} = \frac{2Re[\langle E_H^* \cdot E_V \rangle]}{\langle |E_H|^2 \rangle + \langle |E_V|^2 \rangle} \approx \frac{2E_V}{E_H}.
\]

(28)

Combining Eqs. (26) and (28), the ellipticity of the probe light is no more than
\[
e \leq 2 \times 10^{-3}.
\]

(29)

For Cs D2 transition, the corresponding dipole matrix element in Eq. (17) should be replaced as \( \langle 6S_{1/2}|er|6P_{3/2} \rangle \approx 4.48e_{a_0} \), and the \( J_e = 1/2 \) should be replaced by \( J_e = 3/2 \). The vector light shifts due to the probe light for atomic spin precession frequencies are
\[
\Delta f_{3, \text{prb}, \text{VLS}} \approx -1.42 \times 10^{-3} \text{ Hz},
\]

(30)

\[
\Delta f_{4, \text{prb}, \text{VLS}} < 10^{-6} \text{ Hz}.
\]

(31)

The corresponding upper limit of the systematic errors due to the vector light shift of the probe light is calculated to be
\[
R_{\text{err,prb,VLS}} = 1.174 \times 10^{-7}.
\]

(32)

However, we need to note that, the glass window may be another factor that will change the ellipticity of the probe light when it enters the atomic vapor cell, and therefore increase the systematic errors due to the vector light shift of the probe light. Due to stresses from the joint with the cylinder body which may be created in the manufacturing process, the circular window of the atomic vapor cell could be birefringent. And to suppress the potential increase in ellipticity, the point of incidence for the probe light to enter the atomic vapor cell is fixed at the middle of the plane, where the stresses from the joint should be “neutralized” in each direction.

The tensor light shift of Zeeman sublevel \( |F, m_F \rangle \) induced by the linearly polarized light near Cs D2 line is given by [42]:
\[
\Delta f_{\text{TLS}} = \frac{-\langle |E_0|^2 \rangle}{2} \left\{ \frac{40F(2F + 1)(2F - 1)}{3(F + 1)(2F + 3)} (3\cos^2 \alpha - 1) \right\} \times \left\{ \frac{3m_F^2 - F(F + 1)}{2F(2 - F)} \right\} \left\{ \langle 6S_{1/2}|er|6P_{3/2} \rangle \right\}^2 \mathcal{T}(\nu),
\]

(33)

where \( \alpha \) is the angle between the magnetic field and the probe light polarization, and \( \mathcal{T}(\nu) \) is sum of the imaginary part of three Voigt profiles [38, 39] with dimension Hz\(^{-1}\), given by
\[
\mathcal{T}(\nu) = \sum_{F_e} \left\{ \begin{array}{ccc} 1 & 1 & 2 \\ F & F & E \end{array} \right\} \left\{ \begin{array}{ccc} J & J & 1 \\ F_e & F_e & I \end{array} \right\} (2F_e + 1) \mathfrak{I}(|\nu|),
\]

(34)

where \( F_e \) represents the hyperfine levels in \( 6P_{3/2} \), \( J_e = 3/2 \) for D2 transition. As is shown in Eq. (32), Zeeman sublevels with \( m_F = \pm F \) have the max tensor light shift. In our system, the probe light polarization is fixed at \( \alpha \approx 54.74^\circ \) to make the item \( (3\cos^2 \alpha - 1) \approx 0 \). The errors of \( \alpha \) are \( \Delta \alpha \lesssim 10^{-5} \text{ rad} \)
\[
\Delta(3\cos^2 \alpha - 1) \approx -3\sin 2\alpha \Delta \alpha \approx 2.8 \times 10^{-5} \text{ rad}.
\]

(35)

Combining Eqs. (32), (33) and (34) with the probe light intensity and frequency detuning, the maximum systematic errors induced by tensor light shift in measuring the frequencies are
\[
\Delta f_{3, \text{prb,TLS}} \approx 3.6 \times 10^{-3} \text{Hz},
\]

(36)

\[
\Delta f_{4, \text{prb,TLS}} < 10^{-6} \text{ Hz}.
\]

The negligible frequency shift for MR signal of \( F_g = 4 \) comes from the fact that the probe light is 14 GHz blue-detuned from the center frequency of the Doppler-broadened Cs D2 \( F_g = 4 \rightarrow F_e \) resonance. The upper limit of systematic errors due to the tensor light shift of the probe light is calculated to be
\[
R_{\text{err,prb,TLS}} = 2.95 \times 10^{-7}.
\]

(37)

The dependence of the index \( R \) on the probe light frequency detuning is shown in Fig. 4b. When the probe light is near resonant to Cs D2 \( F_g = 3 \rightarrow F_e \) transitions, the errors are as large as \( 10^{-4} \). As the inset shows, when the probe light frequency detuning is 5.0 ± 0.5 GHz from Cs D2 \( F_g = 3 \rightarrow F_e = 4 \) transition, the index \( R \) fluctuates within \( \pm 4 \times 10^{-7} \).

The power broadening induced by the probe light comes from the process that atoms absorb the probe light, and the broadening can be calculated from the absorption rate [43]
\[
\Gamma_{\text{abs}} = \sum \sigma(\nu)\Phi(\nu), \quad \sigma(\nu) \propto \mathfrak{R}[|\mathcal{I}(\nu)|],
\]

(38)
Table 2  Estimated upper limits on the effects from the magnetic field and light fields of systematic errors on $R_{err}$

| Source of systematic errors and effect on $R_{err}$ | $2.95 \times 10^{-7}$ | $2.16 \times 10^{-7}$ | $1.17 \times 10^{-7}$ | $5.33 \times 10^{-8}$ | $2.13 \times 10^{-8}$ | $1.56 \times 10^{-8}$ |
|---|---|---|---|---|---|---|
| Tensor light shift (probe) | | | | | | |
| Vector light shift (pump) | | | | | | |
| Vector light shift (probe) | | | | | | |
| Geometric phase effect | | | | | | |
| Nonlinear Zeeman effect | | | | | | |
| Asynchronous optical pumping | | | | | | |

where $\Phi(\nu)$ is the total flux of photons of frequency $\nu$, $\sigma(\nu)$ is the the photon absorption cross-section determined by the real part of the Voigt profile $\Re[\nu(\nu)]$ corresponding to the absorption of light with different frequency for atoms. In our system, the probe light is 5 GHz blue-detuned to the center frequency of the Doppler-broadened Cs D2 $F_g = 3 \rightarrow F_e = 4$ resonance, and the probe light absorption rates for atoms in both hyperfine levels $F_g = 3$ and 4 are more than two orders in magnitude slower than relaxation rate. Therefore, the power broadening in the widths of MR signals from the probe light is negligible. Moreover, because of the inappreciable probe light absorption rate, there is no alignment formed in the atomic ensemble. Consequently, the alignment-to-orientation conversion is out of consideration in our comagnetometer system. The dependence of the index $R$ on the probe light power is shown in Fig. 4c. When the probe light power is larger than $\sim 1$ mW, the errors are slightly larger, maybe because of the fitting errors arising from the small amplitudes of the measured rotation signals. When the probe power is larger, the index $R$ fluctuates within $\pm 4 \times 10^{-7}$.

There are many other sources of systematic errors on $R_{err}$, such as the atomic collisions and the earth rotation, but in the comagnetometer system those from the magnetic field and light fields dominate [13]. Therefore, we focus on the systematic errors induced by the magnetic field and (pump and probe) light fields to testify the optimization measures. The estimated upper limits of systematic errors on $R_{err}$ on the effects from the magnetic field and light fields are listed in Table 2, and the overall value due to these effects are estimated to be

$$R_{err,sum} = \sqrt{\sum (R_{err})^2} = 3.89 \times 10^{-7},$$

which is consistent with the measured value (shown in Fig. 4) when the system works under proper conditions.

Due to the single-species scheme, the systematic errors induced by the magnetic field, which come from the geometric phase effect and nonlinear Zeeman effect, contribute about one order in magnitude less than that induced by the probe light field. And the FID mode helps to reduce the systematic errors induced by the pump light field, which come from its vector light shift and the asynchronous optical pumping effect, to about half that induced by the probe light field.

4 Conclusion and discussion

In conclusion, we proposed and implemented an all-optical single-species Cs atomic comagnetometer based on optical FID. With the single-species scheme, the systematic errors induced by the magnetic field variations and gradients can be suppressed in the comagnetometer. Meanwhile, due to the adoption of optical FID signal detection, the comagnetometer system demonstrated a better ability to suppress the systematic errors induced by the pump light field than those based on frequency scanning. At present, the systematic errors in the comagnetometer are dominated by those induced by the probe light field, which we have not made a special effort to control in this work.

With stabilization of the frequency and power of the pump and probe light, the systematic errors induced by the pump and probe light fields can be further reduced. The optimized all-optical single-species Cs atomic comagnetometer based on optical FID signal may have the potential to search for exotic spin-dependent interactions in the future.

Acknowledgements This work was supported by the National Natural Science Foundation of China (NSFC) (Grant Nos. 61571018, 61531003, 91436210), National Science Fund for Distinguished Young Scholars of China (61225003), and National Hi-Tech Research and Development (863) Program.

References

1. D. Budker, D.F.J. Kimball, Optical Magnetometry (Cambridge University Press, New York, 2013)
2. F. Karwacki, Navigation 27, 72 (1980)
3. S. Lamoreaux, J. Jacobs, B.R. Heckel, F.J. Raab, E. Fortson, Phys. Rev. Lett. 57, 3125 (1986)
4. M.E. Limes, D. Sheng, M.V. Romalis, Phys. Rev. Lett. 120, 033401 (2018)
5. M. Safronova, D. Budker, D. DeMille, D.F.J. Kimball, A. Derevianko, C.W. Clark, Rev. Mod. Phys. 90, 025008 (2018)
6. D. Bear, R. Stoner, R. Walsworth, V.A. Kostelecký, C.D. Lane, Phys. Rev. Lett. 85, 5038 (2000)
7. I. Altarev, C.A. Baker, G. Ban, G. Bison, K. Bodek, M. Daum, P. Fierlinger, P. Geltenbort, K. Green, M.G.D. van der Grinten, E. Gutsmiedl, P.G. Harris, W. Heil, R. Henneck, M. Horras, P. Iaydjiev, S.N. Ivanov, N. Khomutov, K. Kirch, S. Kistryn, A. Knecht, P. Knowles, A. Kozela, F. Kuchler, M. Kuźniak, T. Lauer, B. Lauss, T. Lefort, A. Micheldishvili, O. Naviliat-Cuncic, A. Pazgalev, J.M. Pendlebury, G. Petzoldt, E. Pierre, G. Pignol, G. Quéméner, M. Rebetez, D. Rebreyend, S. Roccia, G. Rogel, N. Severins, D. Shiers, Y. Sobolev, A. Weis, J. Zejma, G. Zsigmond, Phys. Rev. Lett. 103, 081602 (2009)
All-optical single-species cesium atomic comagnetometer with optical free induction decay…

8. F. Allmendinger, W. Heil, S. Karpuk, W. Kilian, A. Scharth, U. Schmidt, A. Schnabel, Y. Sobolev, K. Tullney, Phys. Rev. Lett. 112, 110801 (2014)

9. W. Griffith, M. Swallows, T. Loftus, M.V. Romalis, B. Heckel, E. Fortson, Phys. Rev. Lett. 102, 101601 (2009)

10. C. Abel, N.J. Ayres, G. Ban, G. Bisen, K. Bodek, V. Bondar, M. Daum, M. Fairbairn, V.V. Flambaum, P. Geltenbort, K. Green, Phys. Rev. X 7, 041034 (2017)

11. C. Abel, S. Afach, N.J. Ayres, C. A. Baker, G. Ban, G. Bisen, K. Bodek, V. Bondar, M. Burghoff, E. Chane1, Z. Chowdhuri, P.-J. Chiu, B. Clement, C. B. Crawford, M. Daum, S. Emmenegger, L. Ferraris-Bouchet, M. Fertil, P. Flaux, B. Franke, A. Fratangelo, P. Gellenthin, K. Green, W. C. Griffith, M. van der Grinten, Z. D. Grujić, P. G. Harris, L. Hayen, W. Heil, R. Henneck, V. Hélaine, N. Hild, Z. Hodge, M. Horras, P. Iaydjiev, S. N. Ivanov, M. Kasprzak, Y. Kermaidik, K. Kirch, A. Knec1, P. Knowles, H.-C. Koch, P. A. Koss, S. Komposch, A. Kozela, A. Kraft, J. Kremple, M. Kuźniak, B. Lauss, T. Lefort, Y. Lemière, A. Leredde, P. Mohanmurthy, A. Mtchedlishvili, M. Musgrave, O. Naviliat-Cunic, D. Pais, F. M. Piesga, E. Pierre, G. Pignol, C. Plonka-Spehr, P. N. Prashanth, G. Quéméner, M. Rawlik, D. Rebreyend, A. Schnabel, P. Schmidt-Wettlunger, N. Severijns, D. Shi1, R. Tavakoli Dinani, J. A. Thorne, R. Virot, J. Voigt, A. Weis, E. Wursten, G. Wyszynski, J. Zejma, J. Zenner, and G. Zsigmond, Phys. Rev. Lett. 124, 081803 (2020)

12. D.F.J. Kimball, I. Lacey, J. Valdez, J. Swiatlowski, C. Rios, R. Peregrina-Ramirez, C. Moncrieffe, J. Kremer, J. Dudley, C. Sanchez, Ann. Phys. 525, 514 (2013)

13. D.F.J. Kimball, J. Dudley, Y. Li, D. Patel, J. Valdez, Phys. Rev. D 96, 075004 (2017)

14. T. Wu, J.W. Blanchard, D.F.J. Kimball, M. Jiang, D. Budker, Phys. Rev. Lett. 121, 023202 (2018)

15. P. Gomez, F. Martin, C. Mazzinghi, D.B. Orenes, S. Palacios, M.W. Mitchell, Phys. Rev. Lett. 124, 170401 (2020)

16. Z. Wang, X. Peng, R. Zhang, H. Luo, J. Li, Z. Xiong, S. Wang, H. Guo, Phys. Rev. Lett. 124, 193002 (2020)

17. D. Sheng, A. Kabcenell, M.V. Romalis, Phys. Rev. Lett. 113, 163002 (2014)

18. C. Baker, D. Doyle, P. Gellenthin, M. Green, K. Van der Grinten, P. Harris, P. Iaydjiev, S. Ivanov, D. May, J. Pendlebury, J.D. Richardson, D. Shi1, K.F. Smith, Phys. Rev. Lett. 97, 131801 (2006)

19. M. Ledbetter, S. Pustelny, D. Budker, M.V. Romalis, J. Blanchard, A. Pines, Phys. Rev. Lett. 108, 243010 (2012)

20. S. Afach, C. Baker, G. Ban, G. Bisen, K. Bodek, Z. Chowdhuri, M. Daum, M. Fertil, B. Franke, P. Gellenthin, K. Green, M.G.D. van der Grinten, Z.D. Grujić, P.G. Harris, W. Heil, V. Hélaine, R. Henneck, M. Horras, P. Iaydjiev, S.N. Ivanov, M. Kasprzak, Y. Kermaidik, K. Kirch, P. Knowles, H.-C. Koch, S. Komposch, A. Kozela, J. Kremple, B. Lauss, T. Lefort, Y. Lemière, A. Mtchedlishvili, O. Naviliat-Cunic, J.M. Pendlebury, F.M. Piesga, G. Pignol, P.N. Prashant, G. Quéméner, D. Rebreyend, D. Ries, S. Roccia, P. Schmidt-Wettlunger, N. Severijns, A. Weis, E. Wursten, G. Wyszynski, J. Zejma, J. Zenner, G. Zsigmond, Eur. Phys. J. D 69, 225 (2015)

21. G.D. Cates, S.R. Schaefer, W. Happer, Phys. Rev. A 37, 2877 (1988)

22. J.M. Pendlebury, W. Heil, Y. Sobolev, P.G. Harris, J.D. Richardson, R.J. Baskin, D.D. Doyle, P. Gellenthin, K. Green, M.G.D. van der Grinten, P.S. Iaydjiev, S.N. Ivanov, D.J.R. May, K.F. Smith, Phys. Rev. A 70, 032102 (2004)

23. A. Kastler, J. Opt. Soc. Am. 53, 902 (1963)

24. W.E. Bell, A.L. Bloom, Phys. Rev. Lett. 6, 280 (1961)

25. D. Budker, D. Kimball, V. Yashchuk, M. Zolotorev, Phys. Rev. A 65, 055403 (2002)

26. T. Wu, J.W. Blanchard, G.P. Centers, N.L. Figueroa, A. Garon, P.W. Graham, D.F.J. Kimball, S. Rajendran, Y.V. Stadnik, A.O. Sushkov, A. Wickenbrock, D. Budker, Phys. Rev. Lett. 122, 191302 (2019)

27. G. Breit, I. Rabi, Phys. Rev. 38, 2082 (1931)

28. P. Bevington, R. Gartman, Y.V. Stadnik, W. Chalupczak, Phys. Rev. A 102, 032804 (2020)

29. T.G. Tiecke, “Appendix A” in Feshbach Resonances in Ultracold Mixtures of the Fermionic Quantum Gases 6Li and 40K, Dissertation for the Doctoral Degree (2009)

30. D. A. Steck, Rubidium 85 d line data. http://steck.us/alkalidata (revision 2.1.5, 19 September 2012)

31. D. A. Steck, Rubidium 87 d line data. http://steck.us/alkalidata (revision 2.1.4, 23 December 2010)

32. D. A. Steck, Cesium d line data. http://steck.us/alkalidata (revision 2.1.4, 23 December 2010)

33. W. Li, M. Balabas, X. Peng, S. Pustelny, A. Wickenbrock, H. Guo, D. Budker, J. Appl. Phys. 121, 063104 (2017)

34. V. Gerginov, S. Krzyzewski, S. Knappe, J. Opt. Soc. Am. B 34, 1429 (2017)

35. H. Kleinoppen, Atoms, in Ludwig Bergmann and Clemens Schaefer, Constituents of Matter: Atoms, Molecules, Nuclei, and Particles, ed. by R. Wilhelm (Walter de Gryuter, Berlin, 1997)

36. W. Happer, A. Tam, Phys. Rev. A 16, 1877 (1977)

37. Q.Q. Hu, C. Freier, Y. Sun, B. Leykauf, V. Schkolnik, J. Yang, M. Krutzik, A. Peters, Phys. Rev. A 97, 013424 (2018)

38. W. Happer, B.S. Mathur, Phys. Rev. A 102, 063102 (2016)

39. G.A. Costanzo, S. Micalezio, A. Godone, J.C. Camparo, F. Levi, Phys. Rev. A 93, 063404 (2016)

40. M.D. Swallows, T.H. Loftus, W.C. Griffith, B.R. Heckel, E.N. Fortson, M.V. Romalis, Phys. Rev. A 87, 012102 (2013)

41. M. Auzinsh, A. Berzins, R. Ferber, F. Gahbauer, L. Kalvans, A. Mozera, A. Spis, Phys. Rev. A 91, 053419 (2015)

42. S.K. Peck, N. Lane, D.G. Ang, L.R. Hunter, Phys. Rev. A 93, 023426 (2016)

43. S.J. Seltzer, Developments in Alkali-Metal Atomic Magnetochemistry, Dissertation for the Doctoral Degree (Princeton University, Princeton, 2008)

Publisher’s Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.