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Interactions involving optically oriented Na and Cs atoms in the ground state in a Na−Cs mixture. Frequency shifts.

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The article considers the interaction of alkali atoms Na and Cs in a Na−Cs mixture during optical orientation. Such systems are used as working media when creating devices that operate on the principles of optical orientation of atoms and are used for magnetic measurements. In particular, when developing quantum magnetometers, gyroscopes, and magnetoencephalographs. Optically oriented atoms of alkali metals in the working chamber of such devices collide with each other, and the collisions of atoms are accompanied by the known process of spin exchange (i.e., there is an exchange of electron polarization between the colliding atoms). The spin exchange process significantly affects the width of the magnetic resonance line, as well as the frequency shift. In this work, the dependences of the magnetic resonance frequency shifts on temperature in the range $T = 300-400$ K are calculated, caused by collisions of optically oriented atoms for various hyperfine levels of these atoms (the total moment $F = 4$ for Cs atoms and $F = 2$ for Na atoms). A significant difference in the dependence of shifts on temperature for optical orientation of ^{133}Cs or ^{23}Na atoms for different hyperfine levels has been established.

Keywords: frequency shift, optical orientation, magnetic resonance.

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Introduction

Quantum magnetometers based on the principles of optical orientation (OO) of atoms as a „magnetosensitive element“ use the magnetic resonance line (MRL) of optically oriented atoms as a magnetically sensitive element. The accuracy and sensitivity of these devices are affected by the following characteristics: the signal to noise ratio, the LMR shift relative to the exact resonance, as well as magnetic and radio noise. At the optical orientation of alkali atoms, the sources of frequency shift are pump light, collisions of atoms in the volume of the working chamber with each other or with atoms of the buffer gas. Negative effect of the pumping light may be avoided in conditions of „indirect“ optical orientation of atoms, i.e. when atoms of one kind are optically oriented, while atoms of another kind get polarized as a result of collisions with optically oriented atoms. The study will focus on the case when the absorption chamber contains a mixture of alkali Na and Cs atoms, and the frequency shift is induced by collisions between the Cs−Cs, Cs−Na and Na−Na atoms. A similar situation was considered earlier for the K−Cs mixture [1] that is used in so-called co-magnetometers [2–4] together with the ^{39}K−^{85}Rb mixture [5–7].

The measurement accuracy in devices using the principle of OO atoms depends, in particular, on the correspondence of the measured LMR frequency to the value of the constant magnetic field in which the working chamber of the device, containing a mixture of alkali atoms, is located. The paper will consider a situation when the frequency shift is induced by collisions between polarized atoms in the ground state and the working chamber contains the alkali atom mixture. In this case, the pumping light interacts only with atoms of one kind, while the atoms polarized due to collisions between atoms of another kind do not interact with the light and, of course, there are no light shifts.

Thus, the paper considers frequency shifts caused by spin-exchange collisions of identical and different atoms under OO conditions.

1. Collisions with polarization transfer in the Na−Cs mixture

In collisions involving polarized alkali atoms, polarization transfer occurs if one of the collision partners has been pre-polarized during OO. This kind of process is described using the complex spin exchange cross-section [1,8]:

$$\sigma = \bar{\sigma} + i\bar{\bar{\sigma}}. \tag{1}$$

Here, the first term in the expression (real part) is responsible for polarization transfer and the second term (imaginary part) is responsible for frequency shifts in a system of hyperfine and Zeeman atomic levels.

Figure 1 shows a simplified diagram describing the processes taking place during atom OO in spin exchange conditions. Degeneracy in the Zeeman sublevel system is removed by applying the constant magnetic field H_0, however, in field of the order of Earth's field and lower, the Zeeman sublevels of the corresponding hyperfine state

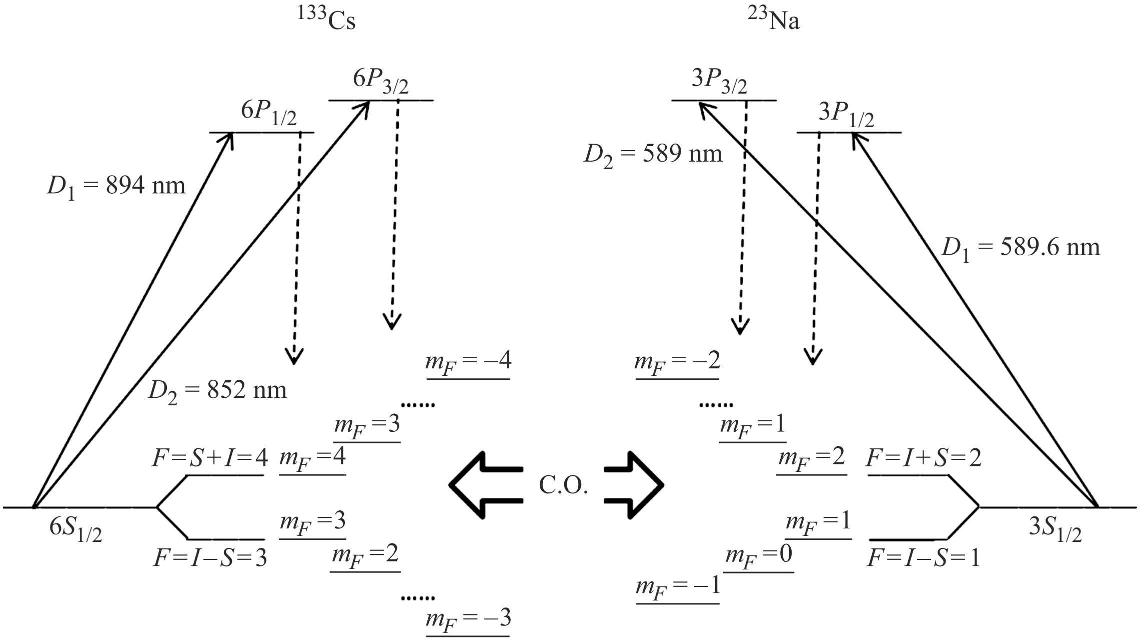

Figure 1. Diagram of lower energy levels (simplified) of alkali ^{133}Cs and ^{23}Na atoms in the presence of the constant magnetic field H_0: D_1 and D_2 are optical resonant line used for atom OO, symbol $\Longleftrightarrow$ indicates that there are spin-exchange collisions between atoms in the ground state, m_F are the total moment projections F on the marked direction (permanent magnetic field direction H_0).

are not allowed. Optical orientation of an alkali atom is performed by the resonance optical radiation of lines D_1 or D_2 (for ^{133}Cs $D_1 = 894\,\text{nm}$, $D_2 = 852\,\text{nm}$, for ^{23}Na $D_1 = 589\,\text{nm}$, $D_2 = 589.6\,\text{nm}$). When using right-handed polarized radiation (σ^+), transitions from the ground $S_{1/2}$-state to the excited $P_{1/2}$ and $P_{3/2}$ states are followed by a change of the total moment projection m_F to $(+1)$, while, when counterclockwise-polarized radiation is used (σ^-) , m_F changes to (-1). Then, decay of the excited state takes place and m_F changes to 0 or ± 1, which leads to nonequilibrium population in the system of Zeeman sublevels of the ground state. Thus polarized kind A alkali metal atoms collide with nonpolarized kind B atoms and transfer a part of their energy to the latter. Note that in collision of alkali atoms at not very low temperatures, i.e. at the temperatures when the hyperfine interaction time $(2\pi\hbar/\Delta E)$ $(\Delta\nu = 1717 \cdot 10^6\,\text{Hz}$ for ^{23}Na and $\Delta\nu = 9192 \cdot 10^6\,\text{Hz}$ for ^{133}Cs [9]) is considerably shorter than the collision time that is approximately 10^{-12} s, the spin exchange process may be considered only as electron spin evolution at the time of collision. In other words, conservation of the total electron spin is expected. At the same time,coupling of the electron and nuclear spins takes place between the collisions. Thus, a molecule formed in collision and consisting of two alkali atoms can be described in its ground state using two potentials corresponding to the total spins of the system $S_1 = 0$ and $S_2 = 1$.

To determine the frequency shift in the Zeeman sublevel system, magnetic resonance is excited by applying a rotating RF field for separation of two signals from two hyperfine states (hyperfine states of the studied atoms have the$g$-factor of the same magnitude and unlike signs) [10]. Thus, the double frequency shift magnitude was determined.

Cross-sections (1) in collisions of the Na–Na, Na–Cs, Cs–Cs atoms have been calcuno commentlated before in [11,12]. The real and imaginary parts of the complex spin exchange cross-section (1) may be expressed through the singlet and triplet terms of the alkali metal dimer. The scattering phases were determined in the Jeffreys quasi-classical approximation modified by Langer [13]. Using of the quasi-classical approximation in the scattering phase calculation is quite justified because, for alkali metal dimers with a large reduced mass μ_{AB} , the centrifugal barrier $\left(\frac{(l+1/2)^2}{2\mu_{\text{AB}}R^2}\right)$ changes just a little as the orbital quantum number l grows compared with the kinetic energy. As a result, contributions of many partial waves shall be considered. Complex spin exchange cross-sections of the studied pairs of atoms were calculated in the collision energy range 10^{-4}–10^{-2} a.u. using the interaction potentials for the singlet and triplet terms of the Na–Cs, Na–Na, Cs–Cs dimers. Then, the Maxwellian averaging of cross-sections over velocities was performed. The calculation results are shown in Table 1.no

Figure 2 shows the dependences of $\bar{\bar{\sigma}}$ of the studied pairs of atoms on temperature as plotted before in [11,12]. It is shown that all cross-sections are within a negative value domain with shift cross-sections for the Na–Na pair differing approximately by half for the Zemke potential [14] (curve *4*) and Zemke–Sovkov potential [14,15] (curve *3*). For curve *4*, the cross-sections were calculated using the singlet and triplet potentials from [14], while for curve *3*, the singlet potential was taken from [14] and the triplet

Table 1. Imaginary parts of the complex spin exchange cross-section $\bar{\bar{\sigma}}(\times 10^{16}\,\text{cm}^2)$ at various temperatures for the Na–Cs, Cs–Cs, Na–Na pairs of atoms

T, K	Na–Cs according to [18]	Cs–Cs	Na–Na according to potentials [12,13]	Na–Na according to potentials [12]
200	−43, 3	−7, 6	−581	−429
250	−45.5	−6.8	−560	−411
300	−47.2	−6.45	−542	−395
350	−48.4	−5.7	−524	−381
400	−49.3	−5.0	−508	−369
450	−49.8	−4.7	−493	−357
500	−50.2	−4.06	−478	−346

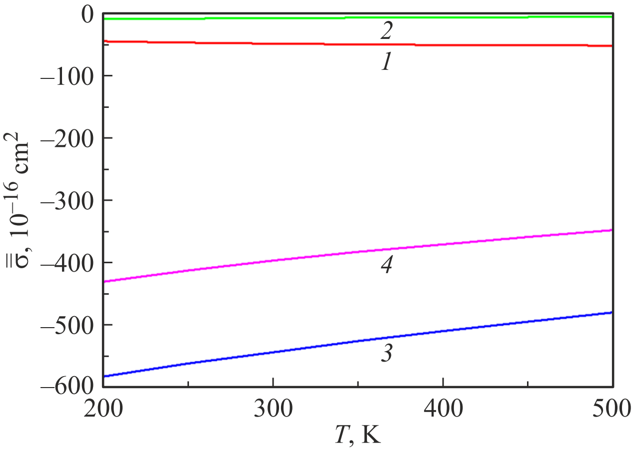

Figure 2. Imaginary part of the complex spin exchange cross-section vs. temperature: *1* — Na–Cs [20], *2* — Cs–Cs, *3* —no comment Na–Na [14,15], *4* — Na–Na [14].

potential was taken from [15]. Negative sign of the shift cross-section indicates that the resonance frequency shifts due to the spin exchange towards lower values with respect to the exact value calculated by $\omega = \gamma H_0$, where γ is the gyromagnetic ratio and H_0 is the permanent magnetic field strength.

2. Frequency shifts

Frequency shifts of two hyperfine sublevels of an alkaline atom are written as [16]:

$$\delta^{(1)}\omega(+) = -\frac{P_z(\mathrm{B})}{2(2I_\mathrm{A}+1)}\left[\bar{\bar{\gamma}}_\mathrm{AB} - \bar{\bar{\gamma}}_\mathrm{AA}\mathrm{B}_-\left(\frac{2I_\mathrm{A}-1}{2I_\mathrm{A}+1}\right)^{1/2}\right], \tag{2}$$

$$\delta^{(1)}\omega(-) = -\frac{P_z(\mathrm{B})}{2(2I_\mathrm{A}+1)}\cdot\left[\bar{\bar{\gamma}}_\mathrm{AB} + \bar{\bar{\gamma}}_\mathrm{AA}\mathrm{B}_+\left(\frac{2I_\mathrm{A}+3}{2I_\mathrm{A}+1}\right)^{1/2}\right]. \tag{3}$$

Frequency shifts $\delta^{(1)}\omega(+)$ of atom A (hyperfine state $F = I + S$, where F is the total atomic moment, I is the nuclear spin, S is the electron spin) and $\delta^{(1)}\omega(-)$ (hyperfine

Table 2. Coefficients B_+ and B_- included in expressions (4) and (5) and depending on the nuclear spins I of the colliding atoms

Nuclear spin, coefficients	^{133}Cs	^{23}Na
I_A	7/2	3/2
B_-	$3\sqrt{5}/4$	$5/6\sqrt{3/2}$
B_+	$7\sqrt{3}/12$	$1/2\sqrt{1/2}$

state $F = I - S$), $P_z(\mathrm{B})$ is the degree of polarization of atom B (atom B is polarized by the resonance optical radiation), $\bar{\bar{\gamma}}_\mathrm{AA}$ is the imaginary part of the spin exchange rate in collision of atoms A. This value depends on the imaginary part of the cross-section $\bar{\bar{\sigma}}_\mathrm{AA}$, mean relative velocity of colliding particles $\langle v_\mathrm{AA}\rangle = (8k_BT/\pi\mu_\mathrm{AA})^{1/2}$ (here k_B is the Boltzmann constant, T is the working chamber temperature, μ_AA is the reduced mass in collision of identicano commentl atoms) and the concentrations N_A of atoms A in the absorption chamber:

$$\bar{\bar{\gamma}}_\mathrm{AA} = N_\mathrm{A}\langle v_\mathrm{AA}\rangle\bar{\bar{\sigma}}_\mathrm{AA}.$$

$\bar{\bar{\gamma}}_\mathrm{AB}$ is written similarly for the case of collision between atoms A and B. Indices A and B are related, respectively, to the Cs or Na atoms.

Coefficients B_- and B_+ in accordance with [16] depend on the nuclear spins of atoms involved in collisions (Table 2). In particular, for the givenno comment pair of Cs and Na atoms, we have

$$\mathrm{B}_+ = \frac{2I_\mathrm{A}+2}{6}\left(\frac{2I_\mathrm{A}+3}{2I_\mathrm{A}+1}\right)^{1/2}, \tag{4}$$

$$\mathrm{B}_- = \frac{2I_\mathrm{A}}{6}\left(\frac{2I_\mathrm{A}-1}{2I_\mathrm{A}+1}\right)^{1/2}. \tag{5}$$

Since expressions (2), (3) include the temperature dependencies of the imaginary parts of the complex spin-exchange rate, then, to calculate the shifts, it is necessary to know the concentration of each of the alkali atoms in the absorption chamber. Concentrations were calculated using the tables from [17]. Since this study addresses a mixture of alkali atoms, then to switch from the absorption chamber temperature to the alkali atom concentration in the chamber it is necessary to use Raoult's law for saturated vapor pressure above the melt of metal mixture. The calculation implied equal weight ratios of alkali metals in the mixture.

Figure 3 shows the dependences of frequency shifts from temperature for the case when Na atoms (B) undergo OO and the frequency shift is observed for Cs atoms (A). It can be seen from the figure that the shift $\delta^{(1)}\omega(-)$corresponding to the hyperfine state of the Cs atom $F = I - S = 3$ is within a positive value domain. According to expression (3) that determines this shift, the major contribution to the shift is made by both terms in square brackets that are defined, in particular, by the imaginary parts of the complex spin exchange rate $\bar{\bar{\gamma}}_{\mathrm{AB}} = \langle \upsilon_{\mathrm{AB}} \rangle N_{\mathrm{AB}} \bar{\bar{\sigma}}_{\mathrm{AB}}$ (for identical atoms, index B is replaced by A). According to the data in Figure 2, $\bar{\bar{\gamma}}_{\mathrm{AB}}$ and $\bar{\bar{\gamma}}_{\mathrm{AA}}$ included in expressions (2) and (3) are defined by the imaginary parts of the complex cross-sections $\bar{\bar{\sigma}}_{\mathrm{AB}}$ and $\bar{\bar{\sigma}}_{\mathrm{AA}}$ that are of like signs. At the same time, $\delta^{(1)}\omega(+)$ for the hyperfine level $F = I + S = 4$ is within a negative value domain and is lower in magnitude than $\delta^{(1)}\omega(-)$. This is due to the facts that according to relation (2) contributions from the first and second terms in the square brackets have unlike signs because of $\bar{\bar{\gamma}}_{\mathrm{AB}} = \langle \upsilon_{\mathrm{AB}} \rangle N_{\mathrm{AB}} \bar{\bar{\sigma}}_{\mathrm{AB}}$ and $\bar{\bar{\gamma}}_{\mathrm{AA}} = \langle \upsilon_{\mathrm{AA}} \rangle N_{\mathrm{AA}} \bar{\bar{\sigma}}_{\mathrm{AA}}$ that in turn depend on $\bar{\bar{\sigma}}_{\mathrm{AB}}$ and $\bar{\bar{\sigma}}_{\mathrm{AA}}$ (Figure 2). The first term depends on the concentration of atoms B, which are the Na atoms is this case, and this concentration is lower than that of the Cs atoms. Thus, interesting us shift is defined by the second term in the square brackets. interaction potential for calculation of the Cs–Cs pair was taken from [18].

During OO, the shift in the Cs atom mixture depends on the imaginary part of the cross-section $\bar{\bar{\sigma}}_{\mathrm{AB}}$ for the Cs–Na pair and the imaginary part of the cross-section $\bar{\bar{\sigma}}_{\mathrm{AA}}$ for the Na–Na pair. Interesting us cross-sections have been calculated before in [11,12] for various interaction potentials. Results for the imaginary parts of cross-sections are shown in Figure 2. As mentioned above, the cross-sections are of like sign and differ approximately by half (Figure 2). A question arises on how such difference will affect the frequency shifts of the Na atoms.

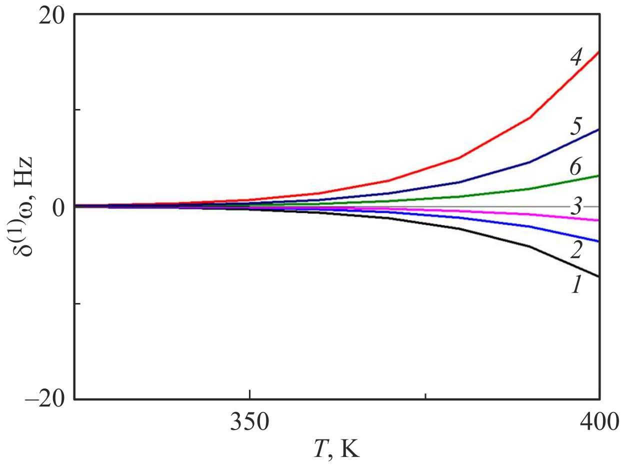

Figure 3. Magnetic resonance frequency shifts vs. temperature for two hyperfine states of the Cs atom $\delta^{(1)}\omega(+)$ (*1–3*) and $\delta^{(1)}\omega(-)$ (*4–6*) at three polarization degrees of the Na atoms: $P_z(\mathrm{B}) = 100$ (*1, 4*), 50 (*2, 5*) 20% (*3, 6*).

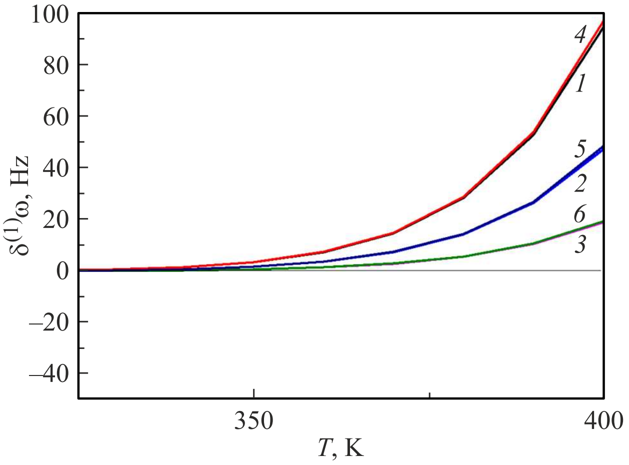

Figure 4. Magnetic resonance frequency shifts vs. temperature for two hyperfine states of the Na atom $\delta^{(1)}\omega(+)$ (*1–3*) and $\delta^{(1)}(-)\omega$ (*4–6*) at three polarization degrees of the Cs atoms: $P_z(\mathrm{B}) = 100$ (*1, 4*), 50 (*2, 5*) 20% (*3, 6*).

Figure 4 shows the frequency shifts vs. temperature for the Na atoms for two hyperfine states during OO of the Cs atoms. The Cs–Na system is described by the potentials from [19], and the interaction between two Na atoms is described by the potentials from [14]. As shown in the figure, frequency shifts for two hyperfine states are of like signs and almost coincide. This is due to the fact that the contribution is made by the first terms in square brackets in expressions (2) and (3) that are defined by collisions between the Na atoms and Cs atoms. As in the previous case, the Cs concentration at the same temperatures is higher than the Na concentration, moreover, the atom collision velocity for the Cs–Na pair is higher than or the Na–Na pair, which defines the magnitudes of $\bar{\bar{\gamma}}_{\mathrm{AB}}$ and $\bar{\bar{\gamma}}_{\mathrm{AA}}$.

The temperature dependences of the frequency shifts for the Na atoms, when the Cs atoms are optically oriented and the Cs–Na system is described by the potentials from [14,15], almost coincide and are of like signs as in the case when the potentials from [14] are used. Therefore, they are not given in this paper.

Conclusion

Note that, despite a significant difference in the cross-sections $\bar{\bar{\sigma}}_{\mathrm{AA}}$ in case when the Cs atom is optically oriented and the shifts are determined for the Na atoms, the frequency shits of these atoms almost coincide. This is due to the fact that the major contribution to the frequency shifts of the Na atoms in the Cs–Na mixture is made by the collisions between the Na atoms and Cs atoms. Moreover, the absolute value of shifts of the Na atoms is more than an order of magnitude higher than the shifts of the Cs atoms.

Conflict of interest

The authors declare that they have no conflict of interest.

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