In view of this situation it would be interesting to determine experimentally the widths of $n+1 \rightarrow n$ lines in hydrogen and hydrogen-like plasmas and at the same time to determine independently the temperature and density. It would be very desirable to measure the widths of several lines in the same experiment which would make it possible to separate the contribution of elastic transitions to the width, which may still be large for $n \sim 10$.

1) The pole potential is used in this case.[4] The regularization parameter introduced in Eq. (1) will be defined later so that the cross sections calculated using Eq. (1) are identical with the Born values.[3]

2) We shall omit the index 1 of the components k_1 and u_1 of the vectors $\mathbf{k}$ and $\mathbf{u}$, denoting them simply by k and u. As in the preceding sections, we shall use again one-dimensional expressions and, therefore, such simplification of the notation should cause no confusion.

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Coherence transfer in metastability exchange in the mixture of the isotopes He^3 and He^4

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We consider coherence transfer in the exchange of metastability in a mixture of He^3 and He^4. It is shown that this process results in appreciable shifts of the magnetic-resonance frequency in the $2\,^3S_1$ states of the He^3 and He^4 atoms. The appearance of these shifts, which has been predicted theoretically, is confirmed by experiments on the optical orientation and magnetic resonance of $2\,^3S_1$-metastable helium atoms. The dependences of these shifts on the temperature, pressure, and concentration of the helium isotopes in the He^3–He^4 mixture are determined.

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INTRODUCTION

In experiments on optical orientation of atoms, the circulation of the coherence between the ground and resonantly excited states leads to the well known optical shift of the magnetic-resonance frequency on account of real optical transitions.[1] The magnitude of this shift is small and is much less than the magnetic-resonance line width, since the light intensity is usually such that the atoms remain in the excited state only a negligible part of the time in comparison with the time of their stay in the ground state.

A similar situation obtains for the helium isotope He^3 when exchange of metastability takes place between two atoms, one of which is in the ground state $1\,^1S_0$ and the other in the metastable $2\,^3S_1$. In this case the atom goes from the ground to the metastable state, and returns to the ground state after a time τ. Coherence is transferred thereby from the ground to the metastable state and back. Since the precession in the metastable state is much faster (the gyromagnetic states for the $2\,^3S_1$ and $1\,^1S_0$ levels, γ_m and γ_f, differ by three orders of magnitude), it follows that an increase of the resonance frequency, comparable in magnitude with the resonance-line width, takes place in the ground state. In view of the practical importance of this question, for example for quantum magnetometers and gyroscopes, this frequency shift had been discussed in many papers,[2–4] where various analytic expressions were given for these shifts. Thus, in[4] the shifts are obtained from the equations for the orientations and alignments of various levels of He^3, and it is noted that these expressions differ from the results of[2,3]. In the same paper,[4] theoretical expressions were obtained for the resonance-frequency shift produced in the metastable state of He^3 by the circulation of coherence between the sublevels $F=1/2$ and $F=3/2$ of the hyperfine structure. Experimental results, on the other hand, were obtained only for the shifts of the NMR frequency in the ground state of He^3.

The present paper is devoted to an investigation of the influence of the coherence circulation on the magnetic resonance of the atoms He^3 and He^4 in the metastable state $2\,^3S_1$ in a mixture of the isotopes He^3 and He^4. The equations for the evolution of the orientations of the different He^3 and He^4 levels were obtained in[5]. At optical orientation and magnetic resonance in the metastable state of the atoms He^3 and He^4, the resonance frequency of the metastable atoms in the mixture of these isotopes should be shifted as a result of exchange of metastability between the atoms of the different isotopes. The excitation energy is transferred in an isotope mixture in a somewhat different manner than in a pure isotope. Thus, an isotope mixture this transfer proceeds partially via the atoms of the other isotope. It is the difference between the gyromagnetic ratios for the $2\,^3S_1$ state of the atoms He^3 and He^4, and consequently the difference between the precession frequencies of the different isotopes in this mixture, which causes the magnetic-resonance frequencies of the metastable atoms to shift, as a consequence of coherence conservation in metastability exchange.

Metastability exchange in helium gives rise to a unique situation, wherein real transitions produce quite appreciable and easily measured and investigated shifts of the magnetic-resonance frequency. This is caused by the very large probability of the metastability-exchange process, inasmuch as one of the partners in the collision is a helium atom in the ground state. It is thus of interest to ascertain the connection between the magnitudes of the resonance-frequency shifts and some quantities of physical interest—the cross sections for metastability exchange between atoms of the same helium isotope or atoms of different helium isotopes.

The paper consists of two parts. In the first, using the equations for the orientations of various He^3 and He^4 levels, we obtain analytic expressions for the resonance-frequency shifts. In the second part we report an experimental check on the obtained theoretical relations, in experiments on optical orientation and magnetic resonance of helium atoms in the $2\,^3S_1$ state in a mixture of He^3 and He^4.

1. THEORETICAL ANALYSIS OF THE CIRCULATION OF COHERENCE AMONG METASTABLE STATES IN AN He^3-He^4 MIXTURE

In experiments on optical orientation of the helium atoms He^3 and He^4, magnetic resonance is produced on one of the $2\,^3S_1$ levels of the metastable state; this leads to coherent precession of the spins in the constant magnetic field H_0. The transfer of coherence from one level to the other occurs in collisions of three types:

$$He^{3*}+He^3\to He^3+He^{2*}\qquad\text{(cross section }\sigma_1\text{)},\tag{1a}$$

$$He^{3*}+He^4\to He^3+He^{4*}\qquad\text{(cross section }\sigma_2\text{)},\tag{1b}$$

$$He^{4*}+He^3\to He^4+He^{3*}\qquad\text{(cross section }\sigma_2\text{)},\tag{1c}$$

where the asterisks mark atoms in the metastable $2\,^3S_1$ state.

If the density of the helium atoms in the absorption chamber is equal to N, and the fraction of the He^3 atoms is αN, then the probabilities per unit time that metastable atoms will take part of one of the processes (1a)–(1c) indicated above are respectively

$$W_1=1/\tau_1=\alpha N v_1\sigma_1,\tag{2a}$$

$$W_2=1/\tau_2=(1-\alpha)Nv_2\sigma_2,\tag{2b}$$

$$W_3=1/\tau_3=\alpha N v_2\sigma_2.\tag{2c}$$

Here v_1 and v_2 are the average relative velocities for He^3-He^3 and He^3-He^4.

A detailed analysis of the influence of these processes on the magnetic resonance in the system He^3-He^4 was carried out in[5], where use was made of a method proposed in[4] for changing over from the equations for the density matrix of the helium-atom system to equations for experimentally observable quantities. This made it possible to describe the evolution of the orientations of the sublevels $He^3-\langle F\rangle_{3/2}$ and $\langle F\rangle_{1/2}$ of the hyperfine structure of He^3, of the metastable state $He^4-\langle S\rangle_{m2}$ of He^4, and of the nuclear orientation of the ground state $He^3-\langle I\rangle_f$ of He^3. From the equations for the orientation components along the constant magnetic fields we obtain the widths of the resonance lines with allowance for the RF field.

To determine the effect of interest to us here, that of the circulation of the coherence on the resonance frequencies, we must resort to the equations for the transverse components of the orientations. Their evolution is determined by the processes (1a)–(1c), and this leads in accord with the data of[5] to the following equations for the transverse components of the orientations of $\langle F_+\rangle_{3/2}$, $\langle F_+\rangle_{1/2}$, $\langle S_+\rangle_{m2}$ and $\langle I_+\rangle_f$:

$$\frac{d}{dt}\begin{pmatrix}\langle F_+\rangle_{3/2}\\ \langle F_+\rangle_{1/2}\\ \langle S_+\rangle_{m2}\\ \langle I_+\rangle_f\end{pmatrix}=[B]\begin{pmatrix}\langle F_+\rangle_{3/2}\\ \langle F_+\rangle_{1/2}\\ \langle S_+\rangle_{m2}\\ \langle I_+\rangle_f\end{pmatrix}.\tag{3}$$

The matrix $[B]$ is of the form

$$[B]=\begin{bmatrix}-\left(\frac{4}{9\tau_1}+\frac{1}{\tau_2}\right)+\frac{2}{3}i\omega_m & \frac{10}{9\tau_1} & \frac{5}{6\tau_3} & \frac{10}{9}\left(\frac{1}{\tau_1}+\frac{1}{\tau_3}\right)\\ \frac{1}{9\tau_1} & -\left(\frac{7}{9\tau_1}+\frac{1}{\tau_2}\right)+\frac{4}{3}i\omega_m & \frac{1}{6\tau_3} & -\frac{1}{9}\left(\frac{1}{\tau_1}+\frac{1}{\tau_3}\right)\\ \frac{2}{3\tau_2} & \frac{4}{3\tau_2} & -\frac{1}{\tau_3}+i\omega_m & 0\\ \frac{1}{3}\left(\frac{1}{T_1}+\frac{1}{T_2}\right) & -\frac{1}{3}\left(\frac{1}{T_1}+\frac{1}{T_2}\right) & 0 & -\left(\frac{1}{T_1}+\frac{1}{T_3}\right)+i\omega_f\end{bmatrix}.\tag{4}$$

Here $1/T_1$, $1/T_2$, and $1/T_3$ are the probabilities of participation in one of the processes (1a)–(1c) for the heli-

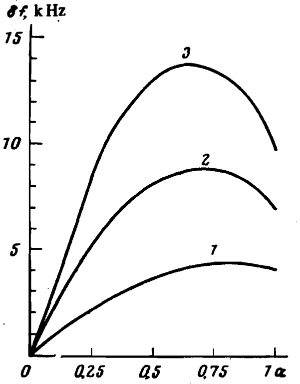

FIG. 1. Dependence of the frequency shift of the magnetic resonance in the metastable state of helium in the He^3–He^4 mixture on the partial pressure of He^3 at a total pressure 0.4 Torr and $H_0 = 1.178$ Oe (curve 1—$T = 125$ K, curve 2—$T = 168$ K, curve 3—$T = 210$ K for the $F = 3/2$ level of He^3.

um atom in the ground $1\,{}^1S_0$ state; $\omega_m = \gamma_m H_0$, $\omega_f = \gamma_f H_0$ are the resonance frequencies in the $2\,{}^3S_1$ state of He^4 and in the ground state of He^3.

The eigenvalues of the matrix $[B]$ determine the position and width of the magnetic-resonance line when the intensity of the RF magnetic field is extrapolated to zero The imaginary parts of these eigenvalues yield the frequencies of the corresponding resonances.[4] The calculation is by successive approximations, the small parameter being $1/\omega_m\tau$. In the zeroth order, i.e., in the absence of metastability exchange ($\tau = \infty$), there is no coherence circulation and the resonance frequencies are equal to the corresponding Larmor precession frequencies in the field H_0. At $\tau \neq \infty$, the coherence in the system of Zeeman sublevels of the state i is transferred in the metastability exchange to the state k, in which the spins precess with a different frequency (ω_k). After a time τ, the coherence can again be transferred to the state i. As a result of this coherence circulation between the levels i and k, the resonant frequency ω_i of the level i either increases when $\omega_k > \omega_i$ or decreases when $\omega_k < \omega_i$. In first order in $1/\omega_m\tau$ the shift of the resonance frequency of the level i as a result of coherence circulation between the levels i and k is determined by the off-diagonal elements b_{ik} and b_{ki} of the matrix $[B]$ and by the difference between the precession frequencies on the levels i and k:

$$\delta\omega_{ik} = \frac{b_{ik}b_{ki}}{\omega_k - \omega_i}. \tag{5}$$

According to this formula, $\delta\omega_{ik}$ is inversely proportional to the frequency difference $(\omega_k - \omega_i)$. This is true in the region $\omega_m\tau \gg 1$, i.e., in sufficiently strong magnetic fields.

We next use this formula with the explicit form of the matrix $[B]$ to calculate the magnetic-resonance frequency shifts for different sublevels of the metastable state He^3 and He^4. No account is taken here of small shifts due to the coherence circulation between the metastable and ground states of He^3 (inasmuch as $1/T_1 \ll 1/\tau_1$).

Magnetic resonance on the $F = 3/2$ *level of* He^3. In this case the resonance frequency increases both when He^4 is added and as a result of the interaction with the atoms on the $F = 1/2$ sublevel. The reason is that the magnetic-moment precession frequencies in the metastable state of He^4 and on the sublevel $F = 1/2$ of He^3 are larger than on the sublevel $F = 3/2$. The frequency shift, according to (5) and (4), is

$$\delta\omega_{3/2} = \frac{5}{27\omega_m\tau_1^2} + \frac{5}{3\omega_m\tau_2\tau_3}. \tag{6}$$

When (2) is taken into account, this expansion becomes

$$\delta\omega_{3/2} = \frac{10}{7}\frac{N^2 v_1^2 \sigma_1^2}{\omega_m}\left[\beta^2\alpha - \alpha^2\left(\beta^2 - \frac{7}{54}\right)\right], \tag{7}$$

where $\beta = \sigma_2/\sigma_1$ is the ratio of the cross sections for metastability exchange in collisions of atoms of different helium isotopes and the same isotope.

With respect to this formula, we can make the following remarks:

1. From the temperature dependences given in[5] for σ_1 and σ_2 it follows that in the temperature interval 77–300 K formula (7) is sufficiently well approximated by a relation of the type $\delta\omega \sim T^{7/2}$, where T is the absolute temperature. Thus, the frequency shift $\delta\omega_{3/2}$ increases sharply with increasing temperature.

2. The frequency shift is a function of the fraction α of the He^3 in the isotope mixture. The shift increases with increasing α, reaches a maximum at $\alpha = \frac{1}{2} + (108\beta^2/7 - 2)^{-1}$, and then decreases to a value corresponding to $\alpha = 1$ (the case of the pure isotope He^3, considered in[4]). The position of the maximum depends on $\beta = \sigma_2/\sigma_1$. Thus, $\alpha_{max} = 0.575$ at $\beta = 1$ and $\alpha_{max} = 0.77$ at $\beta = 1/2$.

3. The frequency shift increases with decreasing constant magnetic field and with increasing atom density in the absorption chamber.

The foregoing regularities are clearly seen in Fig. 1, which shows the frequency shifts of the resonance on the sublevel $F = 3/2$ as functions of α for three temperatures. These regularities were obtained from formula (7) in which σ_1 and σ_2 were taken from[5].

Since the obtained resonance-frequency shifts are large (they reach several dozen kHz) and are commensurate with the resonance-line width, i.e., they can be determined with good accuracy, their measurement may make it possible to determine the cross sections σ_1 and σ_2 in cases when other methods are inconvenient (say at low temperatures).

Magnetic resonance on the sublevel $F = 1/2$ *of* He^3. The circulation of coherence between the level $F = 1/2$ of He^3, on the one hand, and the levels $F = 3/2$ of He^3 and the metastable level of He^4, on the other, leads to a decrease of the resonance frequency, since the precession is slower on the last two levels than on the level $F = 1/2$. The frequency shift is determined according to (5) and (4) by the expression

$$\delta\omega_{1/2} = -\frac{5}{27\omega_m\tau_1^2} - \frac{2}{3\omega_m\tau_2\tau_3}. \tag{8}$$

Substituting here the values of τ_1, τ_2 and τ_3 from (2), we get

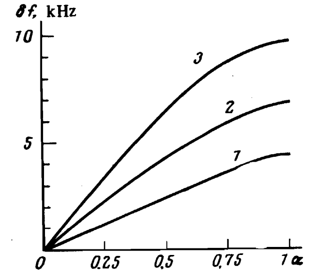

FIG. 2. The same as Fig. 1, but for the $F=1/2$ level of He^3.

$$\delta\omega_{1/2}=\frac{4}{7}\frac{N^2v_1{}^2\sigma_1{}^2}{\omega_m}\left[\beta^2\alpha-\alpha^2\left(\beta^2-\frac{35}{108}\right)\right]. \tag{9}$$

Figure 2 shows the functions $\delta\omega_{1/2}=f(\alpha)$ plotted in accord with formula (9) for three values of the temperature. When N, ω_m, and T are varied, the quantity $\delta\omega_{1/2}$ varies in full analogy with $\delta\omega_{3/2}$, while when α increases this quantity increases monotonically. In this case the frequency shift is smaller than for resonance on the sublevel $F=3/2$.

Magnetic resonance in the metastable state of He^4. Arguments perfectly analogous to those cited above show that the coherence circulation between the $2\,^3S_1$ level of He^4 and the $F=3/2$ level of He^3 leads to a negative frequency shift equal to $-5/3\omega_m\tau_2\tau_3$, while the coherence circulation between the $2\,^3S_1$ level of He^4 and the $F=1/2$ level of He^3 leads to a positive shift equal to $2/3\omega_m\tau_2\tau_3$. The net result is a negative frequency shift

$$\delta\omega_{\mathrm{He}^4}=-\frac{1}{\omega_m\tau_2\tau_3}. \tag{10}$$

We then have according to (2)

$$\delta\omega_{\mathrm{He}^4}=-\frac{6}{7}\frac{N^2v_1{}^2\sigma_1{}^2}{\omega_m}\beta^2\alpha(1-\alpha). \tag{11}$$

Plots of this quantity against α, obtained from formula (11) at three values of T, are shown in Fig. 3. The frequency shift increases sharply with temperature. At all temperatures, the maximum shift corresponds to $\alpha=1/2$. Just as in resonance in the metastable state of He^3, the frequency shift is larger the higher the density of the helium atoms and the weaker the constant magnetic field.

Thus, according to formulas (6)–(11), the frequency shifts at resonance in the metastable state of He^3 and He^4 are connected with the metastability-exchange cross sections and vary with the density of the atoms in the absorption chamber, with the temperature, and with the constant magnetic field. These formulas were obtained in first order in $1/\omega_m\tau=Nv_1\sigma_1/\omega_m$, so that at any given temperature a physically clear picture can be obtained by a suitable choice of the values of N and ω_m. Interest attaches, however, to the influence exerted on the values of the frequency shifts by the terms of higher order in $1/\omega_m\tau$. Calculation of the eigenvalues of the matrix $[B]$ shows that the terms of second order in $1/\omega_m\tau$ influence only the width of the resonance curve, while in third order in $1/\omega_m\tau$ there appears a shift of the resonant frequency of the level i on account of the coherence circulation between the levels i and k. The shift is brought about by the following factors: 1) the damping of the transverse component of the orientation of the level k; 2) the shift of the resonance frequency of the level i in first order in $1/\omega_m\tau$; 3) the appearance of a transverse orientation component of the third level l (which is not directly affected by the coherence circulation).

The correction of third order in $1/\omega_m\tau$ for the resonance frequency of the He^4 atoms in the $2\,^3S_1$ state is equal to

$$\delta\omega_{\mathrm{He}^4}^{(3)}=-\frac{5}{\omega_m{}^3\tau_2{}^2\tau_3{}^2}+\frac{40}{9\omega_m{}^3\tau_1{}^2\tau_2\tau_3}+\frac{15}{\omega_m{}^3\tau_2{}^3\tau_3}+\frac{50}{3\omega_m{}^3\tau_1\tau_2{}^2\tau_3}. \tag{12}$$

The maximum value of this expression is reached at $\alpha=0.222$. The ratio of the shifts given by the terms of first and third order in $1/\omega_m\tau$ is

$$\frac{\delta\omega^{(3)}}{\delta\omega^{(1)}}=-\left(\frac{Nv_1\sigma_1}{\omega_m}\right)^2F(\alpha). \tag{13}$$

Here $F(\alpha)$ is a monotonically decreasing function of α, with particular values $F(0)=7.6$; $F(0.222)=3.3$; $F(1)=-0.21$. According to this formula the third-order corrections are most significant at small values of α and do not exceed several percent at $1/\omega_m\tau\leqslant 0.1$.

The third-order corrections for the resonance frequencies of He^3 are not presented here because the result is too cumbersome. Their calculation is perfectly similar to that for He^4. An estimate, with their aid, of the accuracy of formulas (7) and (9) shows that in this case, too, at $1/\omega_m\tau\lesssim 0.1$ these formulas make it possible to determine α_1 and α_2 with accuracy not worse than 5%.

2. EXPERIMENTAL DETERMINATION OF THE SHIFTS OF THE RESONANCE FREQUENCIES IN THE METASTABLE STATE OF HELIUM IN THE He^3-He^4 ISOTOPE MIXTURE

The experimental setup used in the present study was described in detail earlier.[5]

We used absorption chambers filled with He^3–He^4 mixtures of various proportions, but at a constant pressure 0.4 Torr (at $T=300$ K). The partial pressures of He^3 were equal to 0.1, 0.2, 0.3, and 0.35 Torr. The error in the determination of the pressure was ~8%.

A weak high-frequency gas discharge was excited in the absorption chamber and a fraction (~10^{-5}) of the he-

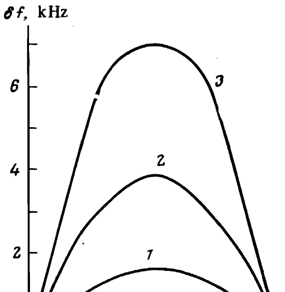

FIG. 3. The same as Fig. 1, but for the level of the $2\,^3S_1$ state of He^4.

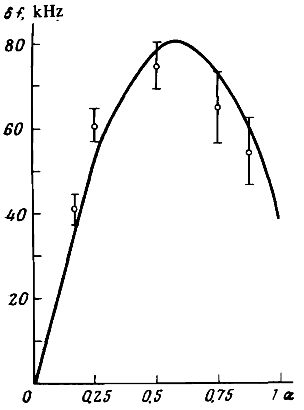

FIG. 4. Dependence of the frequency shift of the resonance of metastable helium atoms at $T=300$ K in an He^3–He^4 mixture on the partial pressure of the He^3 in a field $H_0=1/1/8$ Oe (the solid curve is the theoretical dependence) for the level $F=3/2$ of He^3.

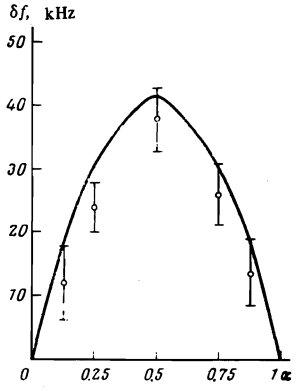

FIG. 5. The same as Fig, 4, but for the $2\,^3S_1$ state of He^4.

lium atoms was raised from the ground $1\,^1S_0$ state to the metastable $2\,^3S_1$ state. The metastable helium atoms were optically oriented in a longitudinal magnetic field with the aid of circularly polarized light of resonant wavelength ($\lambda=10\,830$ Å, $2\,^3S_1 \to 2\,^3P$ transition). The stability of the constant magnetic field was not worse than 2×10^{-4}, corresponding in a field of 1 Oe to a possible frequency drift on the order of 0.6 kHz as a result of the magnetic-field instability. Magnetic resonance was produced in the system of Zeeman sublevels by applying a transverse oscillating magnetic field $H_1 \cos\omega t$. The error in the determination of the frequency depended on the width of the resonance line and on the signal/noise ratio, and ranged from 3.7 to 7.5 kHz.

It is known that in the case of resonance on one sublevel of the metastable state of He^3 nonresonant action of the RF magnetic field on another sublevel causes noticeable shifts of the resonance frequency.[6] To exclude shifts due to the presence of the RF field we therefore extrapolated the position of the center of the resonance line to zero RF-field intensity.

The purpose of the experimental part of the study was to check on the conclusions of Sec. 1. The measurements were made without cooling at $T=300$ K. The constant magnetic field was of the order of 1 Oe; the resonance lines of He^3 and He^4 were then well resolved, notwithstanding their large width at $T=300$ K. Thus, the value of $1/\omega_m\tau$ did not exceed 0.1 and the experimental results can be compared with the theoretical relations derived in first order in $1/\omega_m\tau$.

Figures 4 and 5 show the experimentally obtained values of the magnetic-resonance frequency shifts for the $F=3/2$ level of the $2\,^3S_1$ state of He^3 and for the $2\,^3S_1$ state of He^4 at various partial pressures of He^3 in the He^3–He^4 mixture. The solid curves in the same figures show the theoretical plots of formulas (7) and (11) at $\sigma_1=\sigma_2=7.6\cdot10^{-16}\ \mathrm{cm}^2$[4] and a pressure 0.4 Torr (at $T=300$ K). The calculated theoretical frequency shifts contain a certain constant factor, determined with accuracy ~15% (in accordance with the possible error in the determination of the experimental values of σ and N), which does not influence the character of the dependence of the shift on the partial pressure of He^3. It is seen from Figs. 4 and 5 that the agreement between the experimental results and the theoretical relations is very good.

Thus, the shifts of the magnetic resonance in the metastable state of He^3 and He^4 are quite appreciable and can be reliably measured. As shown above, they can be well interpreted on the basis of the formulas of Sec. 1. This attests to the sufficient completeness and accuracy of the magnetic-resonance information that can be obtained from the equations for the evolution of the orientations of the various metastable-state levels of He^3 and He^4.

Measurement of the shifts of the resonance frequency can be of additional interest for the determination of the metastability-exchange cross sections σ_1 and σ_2 from experiments on optical orientation and magnetic resonance of metastable helium atoms. These quantities are usually obtained from the magnetic-resonance line widths.[4,5] At low temperature, however, the metastability-exchange cross sections decrease sharply and the magnetic-resonance line width is governed to the same degree by the metastability exchange as by relaxation on the walls of the absorption chamber, by the inhomogeneity of the constant and RF magnetic fields, by various processes in the gas discharge, and others.[7] It appears that in such a situation the information obtained from the magnetic-resonance shifts is quite valuable, and sometimes the only reliable one. To obtain appreciable frequency shifts in such cases it is necessary to work with weak magnetic fields at a maximum possible density of the helium atoms in the absorption chamber ($N\sim10^{17}$ cm^{-3}).

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