Effect of nuclear spin on the exchange line shift in optically pumped gases

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It is pointed out that hyperfine interaction should be taken into account in the equation for the spin density matrix, which describes the evolution of atom polarization in optically pumped gases. A dispersion equation is derived for the possible frequencies of atom polarization oscillations in a polarized gas. It is shown that the very possibility of observation of paramagnetic resonance exchange line shifts is due to the presence of hyperfine interaction. An analysis is made of the dependence of the paramagnetic resonance excitation amplitude at frequencies involving an exchange shift of the relation between the energy of hyperfine interaction between the electron and nucleus and the exchange interaction energy.

The influence of the spin of the nucleus on the electron spin resonance line shift due to exchange interaction of electrons and atoms in optically pumped gases has been extensively discussed in the literature of late[1-7]. It is stated in some papers[1-4] that the spin of the nucleus plays no role, and the equation for the density matrix is expressed in a form that does not contain a term describing the hyperfine interaction of the electron and the nucleus. In other papers[5,6], on the other hand, using the interaction of an atom with a buffer spinless gas as an example, it is shown that the hyperfine interaction leads to damping of the longitudinal polarization of the electron of the atom, with two relaxation times. However, no analysis is presented of the influence of the nuclear spin on the shifts of the lines in a polarized gas.

As will be shown below, the very possibility of observing a shift of the ESR line is connected with the existence of a hyperfine interaction between the electron and the nucleus. It will also be shown that allowance for the nuclear spin leads to a change in the expressions describing the relaxation of the polarization of the atomic electrons in comparison with the results given by Balling et al.[1,2].

Let us stop first to discuss the form of the equation for the single-particle density matrix. Since this question is debatable, we derive the equation of interest to us by resorting to the general theory (see, for example,[8]). According to this theory, the density matrix ρ of the system satisfies the equation

$$i\hbar\partial\rho/\partial t=[\mathscr{H},\rho], \tag{1}$$

where $\mathscr{H}=\mathscr{H}_A+\mathscr{H}_B+\mathscr{H}_{AA}+\mathscr{H}_{BB}+\mathscr{H}_{AB}$ is the total Hamiltonian of the entire system and $\mathscr{H}_{A(B)}$ is the Hamiltonian of the noninteracting atoms of type A(B). It obviously contains a term describing hyperfine interaction of the atomic electron with the nuclear spin. $\mathscr{H}_{AA,(BB),(AB)}$ is the part of the total Hamiltonian describing the interaction of the atoms with one another.

Averaging (1) over all variables with the exception of the variables, say, of the atom A, we obtain from (1) an equation for the single-particle spin density matrix of the atom A:

$$i\hbar\frac{\partial\rho(A)}{\partial t}=[\mathscr{H}(A),\rho(A)]+i\hbar\left(\frac{\partial\rho(A)}{\partial t}\right)_{\text{coll}}, \tag{2}$$

where

$$i\hbar(\partial\rho(A)/\partial t)_{\text{coll}}=\mathrm{Sp}'[\mathscr{H}',\rho],\quad \mathscr{H}'=\mathscr{H}-\mathscr{H}(A),$$

and Sp′ denotes averaging over all the variables with the exception of the variables describing the atom of type A.

If we calculate $(\partial\rho/\partial t)_{\text{coll}}$ by the method developed in[9-11], then we can express the collision term $(\partial\rho/\partial t)_{\text{coll}}$ in terms of the atom-atom scattering amplitude. A collision term obtained in this manner coincides in form with the collision term derived in[1-4] and will therefore not be written out here.

Equation (2) differs from the equation for the density matrix in[1-4] in the presence of the term $[\mathscr{H}(A), \rho(A)]$, which describes free motion of the atom with allowance for the hyperfine interaction. In other words, the equation obtained for the density matrix in[1-4] determines only the collision part of the total equation (2).

Thus, from the point of view of general theory, inclusion of the hyperfine interaction in the equation for $\rho(A)$ is essential. It remains only to clarify the role of this interaction. For simplicity, we shall henceforth neglect the level broadening due to collisions. We therefore discard from Eq. (27) given in[1] for $d\rho/dt \equiv (\partial\rho/\partial t)_{\text{coll}}$ all the terms that do not contain the imaginary unit i. As a result we obtain from (2), for atoms with one external electron in the S state, a system of equations in the form

$$\begin{gathered}i\hbar\frac{d\rho(A)}{dt}=[(a_A\mathbf{S}_A\mathbf{j}_A-\gamma\mathbf{S}_A\mathbf{H}-\alpha_{AA}\mathbf{S}_A\mathbf{P}_A-\alpha_{AB}\mathbf{S}_A\mathbf{P}_B),\rho(A)],\\ i\hbar\frac{d\rho(B)}{dt}=[(a_B\mathbf{S}_B\mathbf{j}_B-\gamma\mathbf{S}_B\mathbf{H}-\alpha_{BA}\mathbf{S}_B\mathbf{P}_A-\alpha_{BB}\mathbf{S}_B\mathbf{P}_B),\rho(B)],\end{gathered} \tag{3}$$

where $a_A(a_B)$ is a constant of the hyperfine interaction of the electron with the nucleus in the atom of type A(B), γ is the magnetomechanical ratio of the electron, $j_A(j_B)$ is the operator of the nuclear spin of the atom A(B), H is the external magnetic field, $S_{A(B)}$ is the electron spin operator, $P_A = 2\,\mathrm{Tr}\,S_A\rho(A)$ is the polarization vector of the electron of the atom A, and P_B is the same for the atom B. The terms containing the constants $\alpha_{AA}=\hbar K_{AA}/T_{AA}$ and $\alpha_{BB}=\hbar K_{BB}/T_{BB}$ stem from the exchange scattering of the atoms of type A(B) by one another. An expression for them is given in[1,4]. The terms containing the constants $\alpha_{AB}=\hbar K_{AB}/T_{AB}$ and $\alpha_{BA}=\hbar K_{BA}/T_{BA}$ are the result of the exchange scattering of the atom of type A by the atom of type B. Expressions for them can be found likewise in[1,4].

We note that apart from allowance for the hyperfine interaction, the system (3) differs from the system used to analyze the line shift in[1,2] in that terms describing collisions of like atoms are taken into account. In addition, in the cited papers they used only one equation for the analysis of the line shift, and the polarization of the second system was assumed fixed. It will be shown below that to analyze the shift it is necessary to solve the simultaneous system (3).

It should be noted that Eqs. (3) can be obtained with the aid of the following simple reasoning. As indicated by us earlier[12], owing to the refraction in the polarized gas, the atom is acted upon by an effective magnetic field that stems from the exchange interaction and is analogous in nature to the effective molecular field responsible for ferromagnetism. A similar conclusion was subsequently drawn by Lambert for the case of optical pumping[3]. From the indicated point of view, the system (3) is perfectly understandable: it describes the temporal evolution of the polarization of an atom of type A(B) situated in an effective external magnetic field

$$\mathbf{H}_{\mathrm{eff}}(A)=\mathbf{H}+\frac{\alpha_{AA}}{\gamma}\mathbf{P}_A+\frac{\alpha_{AB}}{\gamma}\mathbf{P}_B \qquad \left(\mathbf{H}_{\mathrm{eff}}(B)=\mathbf{H}+\frac{\alpha_{BA}}{\gamma}\mathbf{P}_A+\frac{\alpha_{BB}}{\gamma}\mathbf{P}_B\right).$$

We proceed now to an analysis of the influence of the hyperfine interaction on the paramagnetic resonance in the system under consideration. We shall show first that when we discard from the equation for ρ the term that describes the hyperfine interaction, the exchange line shift is not observable. In fact, if we discard the indicated term and then multiply the first equation of (3) by $\mathbf{S}_A$ and the second by $\mathbf{S}_B$, and take the trace of both equations, then we obtain

$$\begin{aligned}\frac{d\mathbf{P}_A}{dt}&=-\frac{\gamma}{\hbar}[\mathbf{H}\mathbf{P}_A]-\frac{\alpha_{AB}}{\hbar}[\mathbf{P}_B\mathbf{P}_A],^{*}\\ \frac{d\mathbf{P}_B}{dt}&=-\frac{\gamma}{\hbar}[\mathbf{H}\mathbf{P}_B]-\frac{\alpha_{BA}}{\hbar}[\mathbf{P}_A\mathbf{P}_B].\end{aligned} \tag{4}$$

The system (4) describes the temporal evolution of spins having identical magnetomechanical ratios and coupled with one another by exchange forces. Its form is perfectly analogous to the form of the system of equations describing the temporal evolution of the polarization of two sublattices in an antiferromagnet in the limit of identical magnetomechanical ratios of the spins of both sublattices[13,14]. According to[13,14], in the case of small transverse oscillations of the polarization vector about the stationary direction of the polarization vector in the system (4), there are two natural frequencies

$$\hbar\Omega_1=\gamma H_z,\quad \hbar\Omega_2=\gamma H_z+\alpha_{AB}P_{Az}+\alpha_{BA}P_{Bz}. \tag{5}$$

We note that in[1-4], in which the hyperfine interaction is neglected, the electron spin precession frequency was analyzed using only one equation of (4), while the polarization of the second system was assumed fixed and directed along the z axis. As a consequence it was found that in the system A there is only one natural frequency $\hbar\Omega=\gamma H_z+\alpha_{AB}P_{Bz}$. It will be shown later on that such an expression arises for the frequency only as a result of the hyperfine interaction in sufficiently weak exchange fields.

Thus, polarization oscillations in the system on discarding the term containing the hyperfine interaction occur at two frequencies Ω_1 and Ω_2, and only the second of these contains the contribution of the exchange interaction. However, as shown in[13] (see also[14]), where ferromagnetic resonance in a system of sublattices was considered, the transitions at the frequency Ω_2 are forbidden if the magnetomechanical ratios for the spins participating in the resonance are identical. This is precisely the situation in our case.

Thus, in the absence of interactions between the electron spin and the nuclear spin, the exchange shift in the experiments of Balling et al.[1,2] would be unobservable.

We then take into account in (3) the term $a\mathbf{S}\cdot\mathbf{j}$, which describes the hyperfine interaction, and proceed to a more detailed investigation of its influence on the possibility of exciting resonance at the exchange frequency.

Under stationary conditions, in the absence of an alternating external field $\mathbf{h}(t)$, the polarization vector $\mathbf{P}_{A(B)}=\mathbf{P}_{0A(B)}$, and its direction is either parallel or antiparallel to the constant magnetic field $\mathbf{H}_0$. We choose the direction of $\mathbf{H}_0$ to be the quantization axis z, and then write down the density matrix in the representation of the eigenfunctions $|\,i\,\rangle$ of the Hamiltonian

$$\mathscr{H}_0=a\mathbf{S}\mathbf{j}-\gamma S_z H_{z\,\mathrm{eff}}$$

An expression for the eigenvalues E_i corresponding to the functions $|i\rangle$ is given, for example, in[15].

We note that in the case under consideration, at $\mathbf{h}(t)=0$, the density matrix is diagonal. If we now turn on the field $\mathbf{h}(t)$, then off-diagonal elements appear in the density matrix and the polarization vector of the atoms becomes time-dependent:

$$\mathbf{P}=2\sum_{ik}\langle i|\mathbf{S}|k\rangle\,\rho_{ki}=\mathbf{P}_0+\delta\mathbf{P}(t).$$

In the representation of the eigenfunctions of the Hamiltonian $\mathscr{H}_0$, the system (3) for the off-diagonal elements of the density matrix can be written in the form

$$\begin{aligned}&i\hbar d\rho_{ik}(A)\,/\,dt=(E_i-E_k)\rho_{ik}(A)\\ &+(\rho_{ii}(A)-\rho_{kk}(A))\langle i|\mathbf{S}_A|k\rangle[\alpha_{AA}\delta\mathbf{P}_A(t)+\alpha_{AB}\delta\mathbf{P}_B(t)+\gamma\mathbf{h}(t)],\\ &i\hbar d\rho_{lm}(B)\,/\,dt=(\varepsilon_l-\varepsilon_m)\rho_{lm}(B)\\ &+(\rho_{ll}(B)-\rho_{mm}(B))\langle l|\mathbf{S}_B|m\rangle[\alpha_{BA}\delta\mathbf{P}_A(t)+\alpha_{BB}\delta\mathbf{P}_B(t)+\gamma\mathbf{h}(t)].\end{aligned} \tag{6}$$

In deriving (6) from (3) we have left out the terms proportional to the product of $\delta\mathbf{P}(t)$ by the off-diagonal elements of the density matrix ρ, since they are of second order of smallness in comparison with the retained terms (it is assumed that $|\,\mathbf{h}(t)\ll\mathbf{H}$ and that the deviation of the polarization from the stationary value is small, i.e., $|\,\delta\mathbf{P}(t)\,|\ll\mathbf{P}_0$).

To find the solution of the system (6), we carry out a Fourier transformation with respect to time

$$\rho(t)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\rho(\omega)\,e^{-i\omega t}\,d\omega.$$

Further, solving the system (6) with respect to $\rho_{ik}(A,\omega)$ and $\rho_{lm}(B,\omega)$, multiplying the first of the obtained expressions by $\langle\,k\,|\,\mathbf{S}_A\,|\,i\,\rangle$ and the second by $\langle\,m\,|\,\mathbf{S}_B\,|\,l\,\rangle$, summing over all $i\neq k$, and recognizing that

$$\delta\mathbf{P}=2\sum_{i\neq k}\langle k|\mathbf{S}|i\rangle\,\rho_{ik},$$

we obtain

$$\delta\mathbf{P}_A(\omega)=2\sum_{i\neq k}\frac{\langle k|\mathbf{S}_A|i\rangle(\rho_{ii}-\rho_{kk})}{\hbar\omega-(E_i-E_k)}$$

$$\times[\langle i|\mathbf{S}_A|k\rangle(\alpha_{AA}\delta\mathbf{P}_A(\omega)+\alpha_{AB}\delta\mathbf{P}_B(\omega)+\gamma h(\omega))],$$

$$\delta\mathbf{P}_B(\omega)=2\sum_{l\neq m}\frac{\langle m|\mathbf{S}_B|l\rangle(\rho_{ll}-\rho_{mm})}{\hbar\omega-(\varepsilon_l-\varepsilon_m)} \tag{7}$$

$$\times[\langle l|\mathbf{S}_B|m\rangle(\alpha_{BA}\delta\mathbf{P}_A(\omega)+\alpha_{BB}\delta\mathbf{P}_B(\omega)+\gamma\mathbf{h}(\omega))].$$

It is convenient to examine the system (7) separately for the transverse components of the polarization vector $\delta\mathbf{P} = (\delta P_x, \delta P_y, 0)$ and for the longitudinal component $\delta\mathbf{P} = (0, 0, \delta P_z)$. In the former case we introduce the linear combinations $\delta P_\pm = \delta P_x \pm i\delta P_y$ and $h_\pm = h_x \pm ih_y$. We then get from (7)

$$\begin{gathered}[1-\eta_A(\omega)\alpha_{AA}]\delta P_{A(-)}(\omega)-\eta_A(\omega)\alpha_{AB}\delta P_{B(-)}(\omega)=\eta_A(\omega)\gamma h_-(\omega),\\ -\eta_B(\omega)\alpha_{BA}\delta P_{A(-)}(\omega)+[1-\eta_B(\omega)\alpha_{BB}]\delta P_{B(-)}(\omega)=\eta_B(\omega)\gamma h_-(\omega),\end{gathered} \tag{8}$$

where

$$\eta_A(\omega)=\sum_{i\neq k}\frac{\langle k|S_{A(-)}|i\rangle\langle i|S_{A(+)}|k\rangle(\rho_{ii}-\rho_{kk})}{\hbar\omega-(E_i-E_k)},$$

$$S_{(\pm)}=S_x\pm iS_y,$$

and $\eta_B(\omega)$ is obtained from $\eta_A(\omega)$ by replacing the subscript A by B and replacing E by ϵ. Equations for δP_+ are obtained from (8) by replacing the subscript (-) by (+). Equations for the determination of the longitudinal oscillations δP_z of the polarization vector are obtained by replacing the quantities $S_{(\pm)}$ by $S_z\sqrt{2}$ in the coefficients of η. From (8) we obtain the following expression for $\delta P_{A(-)}(\omega)$:

$$\begin{gathered}\delta P_{A(-)}(\omega)=\chi_A(\omega)h_-(\omega),\\ \chi_A(\omega)=\eta_A(\omega)\gamma\frac{1-\eta_B(\omega)\alpha_{BB}+\alpha_{AB}\eta_B(\omega)}{[1-\eta_A(\omega)\alpha_{AA}][1-\eta_B(\omega)\alpha_{BB}]-\eta_A(\omega)\alpha_{AB}\eta_B(\omega)\alpha_{BA}}\end{gathered} \tag{9}$$

If the system is excited by a transverse alternating magnetic field $\mathbf{h}(t)$ with components $h_x = h_\perp \cos \Omega t$ and $h_y = h_\perp \sin \Omega t$, then $h(t) = h_\perp e^{-i\Omega t}$ and $\mathbf{h}_-(\omega) = 2\pi h_\perp \delta(\omega - \Omega)$. Substituting the indicated expression for $h_-(\omega)$ in (9) and taking the inverse Fourier transform, we obtain ultimately for $\delta P_-(t)$ an expression of the type

$$\delta P_{A(-)}(t)=\chi_A(\Omega)h_\perp e^{-i\Omega t}. \tag{10}$$

The quantity $\chi(\Omega)$ has the meaning of the susceptibility of the system. The values of the frequency Ω at which $\chi(\Omega)$ becomes infinite determine, on the one hand, the natural frequencies of the oscillations in the considered system and, on the other, the frequencies at which paramagnetic resonance takes place. To find them it is necessary to equate the denominator of (9) (the determinant of the system (8)) to zero, i.e.,

$$[1-\eta_A(\omega)\alpha_{AA}][1-\eta_B(\omega)\alpha_{BB}]-\eta_A(\omega)\alpha_{AB}\eta_B(\omega)\alpha_{BA}=0. \tag{11}$$

Knowledge of these natural frequencies (the roots of Eq. (11)) enables us to rewrite (9) in the form

$$\begin{gathered}\chi_A(\omega)=\eta_A(\omega)\gamma[1-\eta_B(\omega)\alpha_{BB}+\alpha_{AB}\eta_B(\omega)]\\ \times\prod_{i\neq k}(\omega-E_{ik})\prod_{l\neq m}(\omega-\varepsilon_{lm})\Big/\prod_f(\omega-\Omega_f),\end{gathered} \tag{12}$$

where Ω_f are the roots of Eq. (11).

We consider now some particular cases.

If the hyperfine interaction between the spins of the electron and the nucleus is equal to zero then, as shown earlier, the exchange line shift is not observable. It can be shown that the same result follows directly from formula (12).

We now take the hyperfine interaction into account. We assume that the energy of the hyperfine interaction is much smaller than the energy of interaction of the electron with exchange field. The eigenvalues of the Hamiltonian of the atom $\mathscr{H}_0$ can in this case be written in the form (see, for example,[15])

$$\begin{gathered}E_i=-\gamma m_s H_{z\,\mathrm{eff}}(A)+a_A m_s m_j,\\ \varepsilon_l=-\gamma m_s H_{z\,\mathrm{eff}}(B)+a_B m_s m_{j'},\end{gathered} \tag{13}$$

where m_s is the magnetic quantum number of the electron and m_j is the magnetic quantum number of the nucleus with angular momentum j.

Since the operators $S_\pm$ that enter in the expression for $\eta(\omega)$ have nonzero matrix elements for transitions with electron spin flip and without a change of the spin of the nucleus, the difference E_{ik} and ϵ_{lm} in $\eta(\omega)$ can be expressed in the form

$$\begin{gathered}E_{ik}\equiv E_{m_s m_j;\, m_s' m_j'}=[-\gamma H_{z\,\mathrm{eff}}(m_s-m_s')+a_A m_j(m_s-m_s')]\,\delta_{m_j m_j'},\\ \varepsilon_{lm}\equiv\varepsilon_{m_s m_{j'},\, m_s' m_{j'}'}=[-\gamma H_{z\,\mathrm{eff}}(m_s-m_s')+a_B m_{j'}(m_s-m_s')]\,\delta_{m_{j'} m_{j'}'};\end{gathered} \tag{14}$$

According to (14), in strong exchange fields the quantities E_{ik} and ϵ_{lm} form a family of equidistant lines, the separation of which is determined by the hyperfine interaction; these lines are grouped about a value corresponding to the Zeeman splitting in the effective field $H_{z\,\mathrm{eff}}$.

We proceed to find the roots of the dispersion equation (11). To this end, we use a method described for the analysis of similar solutions in[16]. Since the positions of the singular points (14) are known, we can conclude from an examination of a plot of (11) that in the limit of strong exchange fields all but two of the roots of (11) lie between the unperturbed roots E_{ik} and ϵ_{lm}, and consequently the distance between them is on the order of the hyperfine splitting. The two roots Ω_1 and Ω_2 are separated from the indicated group by a distance on the order of the value of the splitting in the exchange field, and coincide in magnitude with the frequencies (5).

Assume now that we are exciting the system at the frequency of the electron Zeeman splitting in an ordinary magnetic field. In this case, taking into account the locations of the roots, we can rewrite (12) in the form

$$\chi_A(\omega)=\mathrm{const}\,\gamma/(\omega-\Omega_1), \tag{15}$$

where the constant is of the order of unity.

On the other hand, if we excite our system at one of the exchange frequencies, say Ω_2, then

$$\chi_A(\omega)=\mathrm{const}'\cdot\frac{\gamma}{\omega-\Omega_2}\frac{\Delta}{\Omega_{\mathrm{exc}}}, \tag{16}$$

where const′ is of the order of unity, Δ is the hyperfine splitting, and $\Omega_{\mathrm{exc}} = \gamma H_{z\,\mathrm{eff}}$ is the precession frequency in the effective field.

Thus, in the second case the amplitude of the polarization excitation is smaller by a factor $\Delta/\Omega_{\mathrm{exc}}$ than the amplitude of excitation of the Zeeman precession frequency in an ordinary magnetic field and tends to zero with increasing exchange interaction, or else when the hyperfine interaction Δ tends to zero.

We now investigate the case when the Zeeman splitting of the levels of the atoms in the exchange field is much smaller than the hyperfine splitting. Two cases are possible, in which the external magnetic field is either much smaller or much larger than the hyperfine interaction. We consider in greater detail the first case.

Let the resonance be produced at the Zeeman frequencies (the experimental situation obtaining in[1,2]).

In this case we can neglect in (8) the terms in the denominators of which the difference $E_i + E_k(\epsilon_l - \epsilon_m)$ is of the order of the hyperfine splitting. We assume furthermore for simplicity that only states with total angular momentum $F = j + \frac{1}{2}$ are populated. Recognizing that the operators $S_{(\pm)}$ change the magnetic quantum number by unity and that the energy separations between neighboring levels belonging to a given value of the total angular momentum are identical in the case of weak fields of interest to us ($E_i - E_k = E_{FmF} - E_{F,mF-1} = \hbar\Omega_A$), we can transform the dispersion equation into

$$\left(1-\frac{\alpha_{AA}f_A}{\omega-\Omega_A}\right)\left(1-\frac{\alpha_{BB}f_B}{\omega-\Omega_B}\right)-\frac{\alpha_{AB}\alpha_{BA}f_Af_B}{(\omega-\Omega_A)(\omega-\Omega_B)}=0, \qquad (17)$$

i.e.,

$$(\omega-\Omega_A-\alpha_{AA}f_A)(\omega-\Omega_B-\alpha_{BB}f_B)-\alpha_{AB}\alpha_{BA}f_Af_B=0, \qquad (18)$$

where

$$f_A=\frac{1}{\hbar}\sum_{m_{j+1/2}}\langle m_{j+1/2}-1|S_{A(-)}|m_{j+1/2}\rangle\langle m_{j+1/2}|S_{A(+)}|m_{j+1/2}-1\rangle(\rho_{m_{j+1/2}}-\rho_{m_{j+1/2}-1}),$$

$$f_B=\frac{1}{\hbar}\sum_{m_{j'+1/2}}\langle m_{j'+1/2}-1|S_{B(-)}|m_{j'+1/2}\rangle\langle m_{j'+1/2}|S_{B(+)}|m_{j'+1/2}-1\rangle$$
$$\times(\rho_{m_{j'+1/2}}-\rho_{m_{j'+1/2}-1})$$

(we recall that each index $ik l m$ in Eq. (8) stands for a pair of quantum numbers F and m_F).

It follows from (18) that the system in question has two natural frequencies

$$\omega_{1,2}={}^1/_2[(\Omega_A+\alpha_{AA}f_A)+(\Omega_B+\alpha_{BB}f_B)]$$
$$\pm{}^1/_2\{[(\Omega_A+\alpha_{AA}f_A)-(\Omega_B+\alpha_{BB}f_B)]^2+4\alpha_{AB}\alpha_{BA}f_Af_B\}^{1/2}. \qquad (19)$$

If the difference between the splittings of the levels of of the atom A and of the atom B is much larger than the precession frequency in the exchange field, or more accurately if

$$[(\Omega_A+\alpha_{AA}f_A)-(\Omega_B+\alpha_{BB}f_B)]^2\gg4\alpha_{AB}\alpha_{BA}f_Af_B,$$

then it follows from (19) that

$$\omega_1=\Omega_A+\alpha_{AA}f_A+\frac{\alpha_{AB}\alpha_{BA}f_Af_B}{(\Omega_A+\alpha_{AA}f_A)-(\Omega_B+\alpha_{BB}f_B)},$$
$$\omega_2=\Omega_B+\alpha_{BB}f_B-\frac{\alpha_{AB}\alpha_{BA}f_Af_B}{(\Omega_A+\alpha_{AA}f_A)-(\Omega_B+\alpha_{BB}f_B)}, \qquad (20)$$

i.e., the precession of the atoms A in the exchange field produced by the polarized atoms B can be considered without taking into account the reaction of the precession of the atoms B in the exchange field produced by the atoms A. Treatment of this kind leads to a precession-frequency shift given by the second terms in formulas (12). Under the conditions of the experiments of Balling et al.[1,2], the indicated correction is small (the exchange field in[1,2] is smaller by approximately two orders of magnitude than the external magnetic field). This is precisely why the exchange shift of the Zeeman frequency of the free electrons can be considered in the manner used in[1,2] without taking into account the usual influence of the polarizations of the interacting subsystems, so that a single frequency $\hbar\Omega = \gamma H_z + \alpha_{AB}P_{Bz}$ is obtained. However, if the densities of the gases A and B are increased in experiments of the type of[1,2] by one or two orders of magnitude, then the systems A and B cannot be considered independently, and Eqs. (18) and (19) must be used for the analysis of the possible resonant frequencies.

Let us consider the second case: the exchange interaction is, as before, much weaker than the hyperfine interaction, but the external field is much stronger. The method of solving Eqs. (8)–(11) is analogous to the first case, but the expressions for the energies E_i and ϵ_l are taken for the case of strong fields (formulas (13), and the eigenfunctions of the atoms are chosen to be the eigenfunctions j_z of the operator S_z. An analysis of the solutions obtained in this manner shows that resonance at the exchange frequency exists in the general case of a partially polarized target, and is suppressed only in a fully polarized target.

We proceed now to consider the influence of the nuclear spin on the longitudinal relaxation of the atomic-electron polarization, the dependence of which on the frequency of the applied magnetic field was investigated experimentally in[1,2]. For concreteness, we consider the system investigated in these studies, namely a gas of polarized atoms interacting with electrons. In the general case, the equations describing the motion of the polarization of the atom with allowance for damping are very complicated. However, if the Zeeman splitting is much larger than the level width, the alternating-field frequency is far from the atomic-level splitting frequency, and the polarization of the system is not very high, then the following system of equations[5] can be obtained for the longitudinal polarization of the atoms:

$$dP_{Az}/dt=\varphi_1-a_1P_{Az}+a_{1j}\langle j_z\rangle+a_{1e}P_{ez},$$
$$d\langle j_z\rangle/dt=\varphi_2+a_{jA}P_{Az}-a_j\langle j_z\rangle+a_{je}P_{ez}, \qquad (21)$$

where all the quantities φ and a depend on the nuclear spin j; their explicit form is given in[5] (Eqs. (85) and (86)). The equation describing the motion of the polarization vector of the electrons (atoms with nuclear spin equal to zero) with allowance for damping is much simpler and can be obtained by adding to the system (4) a term in the form $-(P_e - P_A)/T_{ee}$ (for a derivation see[3]):

$$\frac{d\mathbf{P}_e}{dt}=-\frac{\gamma}{\hbar}[\mathbf{H}\mathbf{P}_e]-\frac{K_{ee}}{T_{ee}}[\mathbf{P}_A\mathbf{P}_e]-\frac{\mathbf{P}_{ee}-\mathbf{P}_A}{T_{ee}}. \qquad (22)$$

Recognizing further that under the conditions of the experiment in[1,2] the effective exchange field is much weaker than the constant external magnetic field, we can disregard the influence of the atomic system on the exchange shift of the electron-system levels (see the discussion of (20)), and discard from (22) the transverse polarization components P_{Ax} and P_{Ay}. Solving Eq. (22) for the transverse components, we obtain

$$P_e^{(+)}=\frac{\gamma}{\hbar}P_{ez}H^{(+)}\left(\omega+\frac{\gamma}{\hbar}H_z+\frac{K_{ee}}{T_{ee}}P_{Az}-\frac{i}{T_{ee}}\right)^{-1},$$
$$P_e^{(-)}=P_e^{(+)*}, \qquad (23)$$

where ω is the frequency of the transverse magnetic field, $H^{(\pm)} = H_x \pm iH_y$. Substituting $P_e^{(\pm)}$ in Eq. (22) for the component P_{ez}, and recognizing that $dP_{ez}/dt = 0$ under the stationary conditions, we obtain the following relation between the longitudinal polarizations of the electrons and atoms:

$$P_{ez}=P_{Az}\left\{1-\frac{\gamma^2}{\hbar^2}H_\perp^2\left[\left(\omega+\frac{\gamma}{\hbar}H_z+\frac{K_{ee}}{T_{ee}}P_{Az}\right)^2+\left(\frac{1}{T_{ec}}\right)^2\right]^{-1}\right\}. \qquad (24)$$

Eliminating P_{ez} from the system (21), we have under stationary conditions ($dP_{Az}/dt = d\langle j_z\rangle/dt = 0$) a simple system of two equations, the solution of which for the polarization P_{Az} is given by

$$P_{Az}=\alpha(j)+\beta(j)\frac{\gamma^2}{\hbar^2}H_\perp^2\left[\left(\omega+\frac{\gamma}{\hbar}H_z+\frac{K_{ee}}{T_{ec}}P_{Az}\right)^2+\left(\frac{1}{T_{ee}}\right)^2\right]^{-1}, \qquad (25)$$

where $\alpha(j)$ and $\beta(j)$ are coefficients independent of ω.

Thus, under stationary conditions in weak exchange fields, the functional dependence of P_{Az} on the frequency of the applied field ω is the same as that used in[1,2]. Allowance for the hyperfine interaction leads only to a change in the value of the stationary polarization P_{Az} in comparison with the results of[1,2], and to the appearance of its dependence on the value of the nuclear spin. We note in conclusion that, in accordance with (21), the hyperfine interaction also leads to a dependence of the type (25) on ω for the longitudinal polarization of the nuclear spin. In addition, if we solve the system (6) with respect to the density matrix (compare with the method of obtaining the dispersion equation), multiply the result by the nuclear spin operator j, and take the trace, then we find that the frequency dependence of the transverse polarization of the nucleus also contains information on the magnitude of the exchange interaction. This enables us to study the exchange interaction by investigating the behavior of the nuclear polarization in an optically pumped gas (for example, by studying the angular distribution or the angular correlation of the γ quanta emitted following decay of an excited nucleus). The foregoing pertains, of course, also to such systems as muonium and positronium in a polarized gaseous (condensed) medium.

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*$[\mathbf{HP}_A] \equiv \mathbf{H} \times \mathbf{P}_A$.

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