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Magnetic and mechanical Faraday effects

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Larmor's theorem expresses the analogy between the motion of a charged particle in a homogeneous magnetic field and the motion in a system in uniform rotation. This analogy suggests that effects of a magnetic field may have a rotational counterpart. We discuss the differences and the analogies between the magnetic Faraday effect and the analogous effect induced by a rotation of the sample. A central issue is to what extent the macroscopic rotation of a material system is communicated to the wave functions of the electrons in the system.

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I. INTRODUCTION

According to Larmor's theorem, the dynamics of a particle with charge q and mass m in a homogeneous magnetic field $\mathbf{B}$ is to first order in the field identical to the dynamics of a particle with mass m as viewed in a coordinate frame in uniform rotation with angular velocity

$$\boldsymbol{\Omega}=\frac{q}{2m}\mathbf{B}\ . \tag{1.1}$$

This angular velocity is just the opposite of the precession frequency induced by the magnetic field. The theorem suggests that effects of a magnetic field have rotational analogs. Let us consider the class of magnetic-field effects on the polarization of light traversing a medium. For example, when linearly polarized light propagates through a medium in a direction parallel to the applied magnetic field, the refractive indices for the right-hand and left-hand circular components are different in general, and a rotation of the polarization direction arises. This is the Faraday effect [1]. Magneto-optical phenomena have been important spectroscopic tools for a long time. In particular the Faraday effect has been extensively studied theoretically [2–5]. More recently, in the case of saturating light intensities the Faraday and the closely related Voigt effect have been applied to study the interaction of atoms and light [6], which also instigated some theoretical work [7,8].

The mechanical analog of the Faraday effect is the rotation of the plane of linear polarization induced by a rotation of the sample, with the axis of rotation parallel to the propagation direction of the light. This angular drag of polarization is analogous to the Fresnel drag of the velocity of light through a moving medium [9]. The angular drag has first been predicted by Fermi [10] for a system without dispersion. Much later, Jones [11] has demonstrated the effect in a rotating glass rod. A theoretical treatment allowing for a frequency dependence of the refractive index has been given by Player [12]. The dispersive contribution should be particularly interesting near resonance, where it is much larger than the nondispersive term. A slightly different situation has been discussed more recently by Silverman [13]. He considered rotationally induced optical activity in free atoms in the case where the full setup is rotating, including not only the sample under consideration, but also the light source, the polarizers, and the detectors, as is the case on the rotating earth. As we shall indicate in Sec. IV, his results could be obtained in a more down-to-earth manner by describing the physics as viewed from the inertial frame.

In view of Larmor's theorem, one might expect that results for the magnetic Faraday effect can be carried over to the mechanical case simply by substituting the frequency of rotation for the Larmor precession frequency. In fact, the relation is more complex, as we have argued in a previous paper [14]. In the present paper we discuss the analogy and the differences between the two effects. Section II describes the mechanical effect by adopting the picture of a rotating potential field in which the electronic dynamics takes place. We point out that in the case of zero electron spin the two effects are complementary rather than analogous. Section III gives a unified treatment of both kinds of perturbations on the refractive index of gases. In the case of the magnetic Faraday effect, this treatment clarifies the relation between the various terms in which the effect is commonly separated [4,5,2]. Sections IV and V discuss the mechanical and magnetic effects, respectively, in the case of an atomic gas with nonzero electron spin. Finally Sec. VI discusses both Faraday effects in the case of a gas of diatomic molecules with zero electron spin.

II. COMPLEMENTARITY RELATION

When linearly polarized light propagating in the Z direction traverses an optically active system, and we take the incoming linear polarization as the X direction, the field at the entrance at $Z=0$ can be decomposed in circular polarizations according to

$$\mathbf{E}(0,t)=-(A/\sqrt{2})\,\mathrm{Re}[\mathbf{u}_{+}-\mathbf{u}_{-}]e^{-i\omega t}\ , \tag{2.1}$$

with A the field amplitude, ω the light frequency, and $\mathbf{u}_{+}$ and $\mathbf{u}_{-}$ the spherical unit vectors

$$\mathbf{u}_{\pm}=\mp(\hat{\mathbf{X}}\pm i\hat{\mathbf{Y}})/\sqrt{2}\ . \tag{2.2}$$

After traversing a path length Z through the medium with refractive indices n_+ for the polarization $\mathbf{u}_+$, and n_- for the polarization $\mathbf{u}_-$, the field is given by

$$\mathbf{E}(Z,t)=-(A/\sqrt{2})\,\mathrm{Re}[\mathbf{u}_+e^{in_+\omega Z/c}-\mathbf{u}_-e^{in_-\omega Z/c}]e^{-i\omega t}\ . \tag{2.3}$$

The refractive indices $n_{\pm}$ always refer to the laboratory frame. The linear polarization is rotated over an angle Θ from the X direction towards the Y direction, where Θ is given by

$$\Theta=-\frac{\omega Z}{2c}(n_+-n_-)\ . \tag{2.4}$$

An intuitively simple example of the mechanical Faraday effect arises in the model case of electrons moving in an external rotating potential field. One may think of the field of a crystal, or of the nuclei in an atomic or a molecular gas. The dynamics of the electrons is then best described in the corotating coordinate frame. We temporarily restrict ourselves to the case of electronic states with zero total spin. In the case of a nonrotating system, the dynamics of the electrons is described by a Hamiltonian H_0, which is the sum of the kinetic energy of the electrons, and a potential-energy term, which contains the Coulomb interactions of the electrons with each other and with the nuclei. For a crystal, when the motion of the nuclei is negligible, the potential energy is independent of time. In the case of a gas, the potential energy has a time dependence corresponding to the thermal motion of the gas particles. The effect of the macroscopic rotation with angular velocity $\mathbf{\Omega}=\Omega\hat{\mathbf{Z}}$ is described by adding to the Hamiltonian H_0 the Coriolis term

$$H_{\mathrm{mech}}=-\Omega L_Z\ , \tag{2.5}$$

with $\mathbf{L}$ the total orbital angular momentum of the electrons. The Hamiltonian H_0+H_{mech} describes the dynamics of the electrons as viewed from the rotating frame, when we assume that the macroscopic rotation is simply superimposed on the thermal motion of the nuclei. In the case of a gas, this assumption is justified, provided that the gas particles have fully accommodated to the rotating wall of the vessel.

The Coriolis term (2.5) modifies the dynamics of the electrons as compared with the case of a nonrotating system. Obviously, this term has the same form as a Zeeman Hamiltonian. In the present case of zero spin, the effect of a uniform magnetic field $\mathbf{B}=B\hat{\mathbf{Z}}$ is described by the perturbation Hamiltonian

$$H_{\mathrm{mag}}=\frac{eB}{2m}L_Z\ , \tag{2.6}$$

with $-e$ the electron charge. (We neglect the diamagnetic term in the Hamiltonian, which is of second order in B.) Hence the Coriolis term (2.5) can be exactly compensated by imposing on the rotating system a uniform magnetic field which has Ω as its Larmor frequency, so that

$$B=\frac{2m}{e}\Omega\ . \tag{2.7}$$

For such a field the two perturbations (2.5) and (2.6) cancel, so that the dynamics of the electrons in the rotating frame and in the magnetic field is described by the Hamiltonian H_0 [15]. The field compels the electronic wave functions to follow the rotation rigidly. Hence the dielectric response of the system to an applied electromagnetic field as viewed in the rotating frame is described by the dielectric susceptibility of the nonrotating medium.

In fact, the derivation of Player of the mechanical Faraday effect was based upon the assumption that in the rotating frame the dielectric response was the same as for the medium at rest [12]. This implied, as pointed out by Baranova and Zel'dovich [16], that he neglected the effect of the Coriolis term (2.5). This assumption of fully rigid rotation leads to the expressions for the refractive indices for circular polarizations [12]:

$$n_{\pm}(\omega)=n_0(\omega)\mp\Omega\frac{dn_0}{d\omega}\mp\frac{\Omega}{\omega}\left[n_0(\omega)-\frac{1}{n_0(\omega)}\right]\ , \tag{2.8}$$

where n_0 is the refractive index of the unperturbed medium. Equation (2.4) then yields

$$\Theta_{\mathrm{rig}}=\frac{\Omega Z}{c}\left[n_0(\omega)-\frac{1}{n_0(\omega)}+\omega\frac{dn_0}{d\omega}\right]\ . \tag{2.9}$$

The derivatives in (2.8) and (2.9) arise from the fact that in the rotating frame the frequencies of circularly polarized light with polarizations $\mathbf{u}_{\pm}$ are observed with frequency shifts $\mp\Omega$. The term proportional to $n_0-n_0^{-1}$ in (2.8) and (2.9) represents the pure kinematic polarization drag due to the rotary motion of the gas.

Since the effects upon the polarization plane due to macroscopic rotation and due to a magnetic field are additive in the linear approximation, we can think of Player's value as consisting of two contributions. One is the true mechanical Faraday effect, and the other the magnetic effect caused by the fictitious field (2.7). Hence we write

$$\Theta_{\mathrm{rig}}=\Theta_{\mathrm{mech}}(\Omega)+\Theta_{\mathrm{mag}}(2m\Omega/e)\ . \tag{2.10}$$

Turning the argument around, we recover the result of Baranova and Zel'dovich [16], who state that a Coriolis term corresponding to a magnetic Faraday effect must be subtracted from Player's result to obtain the correct mechanical effect.

Equation (2.10) implies that the two effects are complementary, in the sense that their sum is fully determined by the unperturbed refractive index, and independent of the details of the perturbations. A nearly spherically symmetric potential implies almost unhindered precession in the case of a magnetic field, and thereby a large magnetic Faraday effect. But this also causes a strong effect of the Coriolis term, which reflects strong slip, leading to a large cancellation of the dispersion term of Θ_{rig} in (2.9), and to a diminished mechanical effect. On the other hand, in a strongly anisotropic potential, the Coriolis term or the Zeeman term has a diminished effect. In the case of rotation, the slip is diminished, and the electronic wave functions are dragged along with the rotation. Then a larger fraction of the dispersive term remains for the mechanical effect, whereas the magnetic effect is diminished.

The magnetic effect is commonly expressed in the form

$$\Theta_{\text{mag}}=VBZ\ . \tag{2.11}$$

In the simple case of a gas with optically active atoms and a light frequency driving a transition between singlet states, the so-called Verdet constant is given by [2]

$$V=\frac{\omega}{c}\frac{e}{2m}\frac{dn_0}{d\omega}\ . \tag{2.12}$$

This relation was already derived by Becquerel long before the advent of quantum mechanics [17]. If we substitute (2.9), (2.11), and (2.12) into (2.10), while taking the field B from (2.7), we find that the dispersive term in (2.9) is exactly canceled by Θ_{mag}. We obtain the simple result

$$\Theta_{\text{mech}}=\frac{\Omega Z}{c}\left[n_0(\omega)-\frac{1}{n_0(\omega)}\right]\ . \tag{2.13}$$

Hence only the kinematic Fermi polarization drag remains for a singlet transition in a dilute monatomic gas. This result is understandable, since as viewed in the laboratory frame the atoms simply follow rectilinear trajectories, and the atomic response to the light is not affected by the rotation. As viewed from the rotating frame, the electronic wave functions will show complete slip.

On the other hand, for a molecular gas, one expects that the rotation of the vessel is transferred to the nuclei. Since the electronic wave functions are coupled to this rotation by nonspherical interactions, they are expected to follow this rotation, giving rise to a mechanical Faraday effect. However, this conclusion should be handled with care, since it is based on a classical picture of molecular rotation. At frequencies where the derivative of the refractive index is appreciably affected by the rotational band structure, this conclusion is not justified. We shall come back to this issue in Sec. VI.

In the remainder of this paper we shall explore the limits of validity of this complementarity picture.

III. POLARIZATION-DEPENDENT REFRACTIVE INDEX OF PERTURBED SYSTEMS

We consider a gas of particles irradiated by a radiation field with frequency ω and polarization vector $\mathbf{u}$. The internal dynamics of the particles is described by a Hamiltonian

$$H=H_0+H_1\ , \tag{3.1}$$

which is separated into the unperturbed part H_0 and a perturbation H_1. We shall be interested in perturbations resulting from a magnetic field or from macroscopic rotation, where H_1 takes a form similar to (2.6) or (2.5). Both perturbations have an axial symmetry. In order to evaluate the rotation of the plane of linear polarization of light propagating along the symmetry axis, we need a general expression for the refractive index of the gas as a function of frequency and polarization. This index is determined by the dispersive part of the dynamic polarizability of the particles. We shall evaluate this index to first order in the perturbation and in the absence of saturation.

The radiation field couples a band or a multiplet G of the ground electronic level to a band E of excited states. The width of all the lines in the band is given by the same linewidth [half width at half maximum (HWHM)] γ. The strength of the absorption is determined by the electric dipole operator $\boldsymbol{\mu}$, and we need only its part $\boldsymbol{\mu}_{EG}$ coupling the substates of the ground level to the substates of the excited level. The Liouville operator $\mathcal{L}$ is defined by specifying its action on a density matrix ρ of a gas particle as

$$i\mathcal{L}\rho=(i/\hbar)[H,\rho]\ . \tag{3.2}$$

We assume that in thermal equilibrium only the ground level is populated, so that the normalized density matrix σ of the gas particles is given by a Boltzmann distribution of the form

$$\sigma=Z_G^{-1}\exp[-\beta H_G]\ , \tag{3.3}$$

where H_G is the Hamiltonian H restricted to the ground level, and where $\beta^{-1}=k_BT$. Note that H_G is defined to have nonvanishing matrix elements only between substates $|g\rangle$ of the ground level, and that $\boldsymbol{\mu}_{EG}$ has only matrix elements between substates $|e\rangle$ and $|g\rangle$ of the excited level and the ground level.

When the gas is traversed by a radiation field with frequency ω and wave vector $\mathbf{k}$, it experiences an electric field

$$\mathbf{E}(\mathbf{r},t)=\operatorname{Re}\mathbf{E}_0(\mathbf{r})e^{-i\omega t}\ . \tag{3.4}$$

The dielectric polarization $\mathbf{P}(\mathbf{r},t)$ equals the particle density N multiplied by the expectation value of the electric dipole of a single particle. Assuming linear response, and adopting the rotating-wave approximation, we obtain

$$\mathbf{P}(\mathbf{r},t)=\operatorname{Re}\mathbf{P}_0(\mathbf{r})e^{-i\omega t}\ , \tag{3.5}$$

with

$$\mathbf{P}_0(\mathbf{r})=N\operatorname{Tr}\boldsymbol{\mu}_{GE}\frac{i/\hbar}{\gamma-i(\omega-\mathcal{L})}[\boldsymbol{\mu}_{EG}\cdot\mathbf{E}_0(\mathbf{r})\sigma]\ . \tag{3.6}$$

Note that the Liouville operator $\mathcal{L}$ operates on optical coherences, so that its eigenvalues are optical transition frequencies. Equation (3.6) specifies the frequency-dependent susceptibility tensor χ, defined by

$$\mathbf{P}_0(\mathbf{r})=\epsilon_0\chi(\omega)\cdot\mathbf{E}_0(\mathbf{r})\ , \tag{3.7}$$

with ϵ_0 the permittivity of the vacuum. This susceptibility would simply be a scalar for an isotropic system, and it is an axially symmetric tensor in the presence of the perturbation H_1. Therefore its eigenvectors must be the two circular polarization vectors $\mathbf{u}_\pm$, given in (2.2), and the unit vector $\hat{\mathbf{Z}}$ in the Z direction. Hence a light beam propagating along the Z direction with circular polarization induces a dipole density $\mathbf{P}$ with the same polarization, and the susceptibility for the light with polarization $\mathbf{u}_\pm$ is simply the matrix element $\mathbf{u}_\pm^*\cdot\chi(\omega)\cdot\mathbf{u}_\pm$ of the tensor χ. The refractive indices $n(\omega,\mathbf{u}_\pm)$ for circularly polarized light are related to the susceptibility by

$$n(\omega,\mathbf{u}_\pm)-1=\tfrac{1}{2}\,\mathrm{Re}\,\mathbf{u}_\pm^*\cdot\chi(\omega)\cdot\mathbf{u}_\pm \ . \tag{3.8}$$

Combination of (3.6)–(3.8) leads to the result

$$n(\omega,\mathbf{u}_\pm)-1=\frac{N}{2\epsilon_0}\,\mathrm{Re\,Tr}\,\mathbf{u}_\pm^*\cdot\boldsymbol{\mu}_{GE}\times\frac{i/\hbar}{\gamma-i(\omega-\mathcal{L})}(\boldsymbol{\mu}_{EG}\cdot\mathbf{u}_\pm\sigma) \ . \tag{3.9}$$

In the simple case of two-state systems the matrix character of σ and $\boldsymbol{\mu}$ disappears, and $\mathcal{L}$ takes the value of the transition frequency. Then one recognizes in (3.9) the well-known dispersion shape.

The separation (3.1) of the Hamiltonian gives rise to a corresponding separation of the Liouville operator

$$\mathcal{L}=\mathcal{L}_0+\mathcal{L}_1 \ , \tag{3.10}$$

and of the ground-state Hamiltonian

$$H_G=H_{G0}+H_{G1} \ . \tag{3.11}$$

This separation (3.11) leads to a perturbative expansion of the ground-state density matrix, which we express to first order as

$$\sigma=\sigma_0+\sigma_1 \ . \tag{3.12}$$

Accordingly, we obtain a first-order contribution to the refractive index from $\mathcal{L}$ in the denominator of (3.9), and a contribution from the ground-state density matrix. The contribution from σ_1 arises from the modification of the populations or of the wave functions of the substates of the ground state. Up to first order we obtain

$$n(\omega,\mathbf{u}_\pm)=n_0(\omega)+n_1(\omega,\mathbf{u}_\pm) \tag{3.13}$$

with

$$n_0(\omega)-1=\frac{N}{2\epsilon_0}\,\mathrm{Re\,Tr}\,\mathbf{u}^*\cdot\boldsymbol{\mu}_{GE}\frac{i/\hbar}{\gamma-i(\omega-\mathcal{L}_0)}(\boldsymbol{\mu}_{EG}\cdot\mathbf{u}\sigma_0) \ , \tag{3.14}$$

the refractive index in the absence of the perturbation with arbitrary polarization $\mathbf{u}$. The first-order correction $n_1(\omega,\mathbf{u}_\pm)$ is the sum of two contributions, corresponding to the modified evolution and to the modified ground-state density matrix, and we find

$$\begin{aligned}n_1(\omega,\mathbf{u}_\pm)=&\frac{N}{2\epsilon_0}\,\mathrm{Re\,Tr}\,\mathbf{u}_\pm^*\cdot\boldsymbol{\mu}_{GE}\frac{i/\hbar}{\gamma-i(\omega-\mathcal{L}_0)}(-i\mathcal{L}_1)\\&\times\left[\frac{1}{\gamma-i(\omega-\mathcal{L}_0)}(\boldsymbol{\mu}_{EG}\cdot\mathbf{u}_\pm\sigma_0)\right]\\&+\frac{N}{2\epsilon_0}\,\mathrm{Re\,Tr}\,\mathbf{u}_\pm^*\cdot\boldsymbol{\mu}_{GE}\\&\times\frac{i/\hbar}{\gamma-i(\omega-\mathcal{L}_0)}(\boldsymbol{\mu}_{EG}\cdot\mathbf{u}_\pm\sigma_1) \ .\end{aligned} \tag{3.15}$$

In both the Coriolis and the Zeeman cases the perturbation H_1 changes sign at a mirror reflection about a plane through the axis. This leads to the symmetry relation

$$n_1(\omega,\mathbf{u}_-)=-n_1(\omega,\mathbf{u}_+) \ . \tag{3.16}$$

Hence it is sufficient to evaluate n_1 for the polarization $\mathbf{u}_+$.

Equation (3.15) is a convenient starting point for an explicit evaluation of the Faraday effect. In order to facilitate the comparison of this general result with the literature [2–5], we shall give an expression for the correction n_1 to the refractive index in terms of matrix elements of the electric dipole operator and the perturbation H_1. The unperturbed eigenstates of the Hamiltonian H_0 obey the eigenvalue equations

$$H_0|g\rangle=E_g|g\rangle, \quad H_0|e\rangle=E_e|e\rangle \ . \tag{3.17}$$

The perturbation H_1 may be separated according to

$$H_1=H_d+H_n \ , \tag{3.18}$$

where H_d is the part of H_1 that is diagonal in the unperturbed energies, whereas H_n is the nondiagonal part of H_1. Hence H_d has nonvanishing matrix elements $\langle e|H_d|e'\rangle=\langle e|H_1|e'\rangle$ and $\langle g|H_d|g'\rangle=\langle g|H_1|g'\rangle$ only when $E_e=E_{e'}$, $E_g=E_{g'}$, whereas H_n has nonvanishing matrix elements only between unperturbed states with different energies. This means that H_d is the part of H_1 that commutes with H_0, whereas H_n is the noncommuting part of H_1. Physically speaking, H_d gives a perturbation of the energy levels E_e and E_g without perturbing the states, whereas H_n perturbs the states $|e\rangle$ and $|g\rangle$. Without loss of generality, we can take the basis of unperturbed eigenstates $|e\rangle$ and $|g\rangle$ that diagonalizes H_d.

The separation (3.18) yields a corresponding separation of the first-order density matrix

$$\sigma_1=\sigma_d+\sigma_n \ , \tag{3.19}$$

whereas the zeroth-order density matrix is

$$\sigma_0=Z_0^{-1}\exp(-\beta H_{G0}) \ . \tag{3.20}$$

The diagonal part of σ_1 is then

$$\sigma_d=-\beta H_d\sigma_0 \ . \tag{3.21}$$

Since the total density matrix (3.3) commutes with the total ground-level Hamiltonian H_G, we find the first-order identity

$$[H_{G0},\sigma_1]=-[H_{G1},\sigma_0] \ . \tag{3.22}$$

The matrix elements of the nondiagonal part σ_n are obtained by taking the matrix elements of (3.22), which gives

$$\langle g|\sigma_n|g'\rangle=Z_0^{-1}\frac{e^{-\beta E_g}-e^{-\beta E_{g'}}}{E_g-E_{g'}}\langle g|H_n|g'\rangle \ . \tag{3.23}$$

The term σ_d gives the correction to the population of the unperturbed states, and σ_n, which only has matrix elements between states with different unperturbed energy, describes the perturbative mixing of the energy eigenstates. In the limit that E_g approaches $E_{g'}$, Eq. (3.23) takes the same form as the matrix elements of (3.21).

The unperturbed refractive index n_0, given in (3.14), can be expressed in terms of matrix elements between the unperturbed states, with the result

$$n_0(\omega)-1=\frac{N}{2\epsilon_0}\sum_g\sum_e \mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_{eg})}Z_0^{-1}\exp(-\beta E_g)\times\langle g|\mathbf{u}^*\cdot\boldsymbol{\mu}_{GE}|e\rangle\langle e|\mathbf{u}\cdot\boldsymbol{\mu}_{EG}|g\rangle \,. \tag{3.24}$$

We now wish to link the result (3.15) to the separate terms A, B, and C, in which Serber describes the magnetic Faraday effect [4,2]. The A term results from the perturbation of the transition frequencies, the B term originates in the mixing of the unperturbed energy eigenstates, and the C term expresses the effect of the modified Boltzmann population of the states. Accordingly, we express the first-order contribution (3.15) as the sum of three terms. The first term results from the contribution of the diagonal part of the perturbation $\mathcal{L}_1$ to the first term on the right-hand side of (3.15). It takes the form

$$n_a(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_g\sum_e \mathrm{Re}\frac{1/\hbar^2}{[\gamma-i(\omega-\omega_{eg})]^2}Z_0^{-1}\exp(-\beta E_g)\times\langle g|\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{GE}|e\rangle\langle e|\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}|g\rangle[\langle e|H_d|e\rangle-\langle g|H_d|g\rangle] \,. \tag{3.25}$$

This term originates from the modification of the transition frequencies $\omega_{eg}=(E_e-E_g)/\hbar$ by the perturbation.

The second contribution to n_1 arises from all nondiagonal terms in (3.15), contained in the nondiagonal contribution to $\mathcal{L}_1$ in the first term, and in the contribution σ_n to σ_1 in the second term on the right-hand side of (3.15). The trace is taken explicitly with the unperturbed states as the basis, and the various terms are combined by using partial fraction expansions such as

$$\frac{1}{\gamma-i(\omega-\omega_{eg})}\,\frac{1}{\gamma-i(\omega-\omega_{e'g})}=\frac{\hbar/i}{E_{e'}-E_e}\left[\frac{1}{\gamma-i(\omega-\omega_{eg})}-\frac{1}{\gamma-i(\omega-\omega_{e'g})}\right] . \tag{3.26}$$

The result is

$$\begin{aligned}n_b(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_g\sum_e \mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_{eg})}Z_0^{-1}\exp(-\beta E_g)\\ \times\Bigg\{\sum_{e'}[\langle g|\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{GE}|e'\rangle\langle e'|H_n|e\rangle\langle e|\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}|g\rangle\\ +\langle g|\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{GE}|e\rangle\langle e|H_n|e'\rangle\langle e'|\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}|g\rangle]\frac{1}{E_e-E_{e'}}\\ +\sum_{g'}[\langle g|H_n|g'\rangle\langle g'|\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{GE}|e\rangle\langle e|\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}|g\rangle\\ +\langle g|\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{GE}|e\rangle\langle e|\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}|g'\rangle\langle g'|H_n|g\rangle]\frac{1}{E_g-E_{g'}}\Bigg\} .\end{aligned} \tag{3.27}$$

This term arises from the perturbation of the energy eigenstates.

The third correction term is due to the diagonal part σ_d of the first-order density matrix σ_1 in (3.15):

$$n_c(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_g\sum_e \mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_{eg})}Z_0^{-1}\exp(-\beta E_g)\times\langle g|\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{GE}|e\rangle\langle e|\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}|g\rangle\times(-\beta\langle g|H_d|g\rangle) \,. \tag{3.28}$$

This term expresses the modification of the thermal substate populations, which results from the splitting of the ground-state level induced by the perturbation.

The total first-order correction to the refractive index is

$$n_1(\omega,\mathbf{u}_+)=n_a(\omega,\mathbf{u}_+)+n_b(\omega,\mathbf{u}_+)+n_c(\omega,\mathbf{u}_+) \,. \tag{3.29}$$

When this sum is substituted into the expression (2.4) for the rotation angle, we obtain in the case of the magnetic Faraday effect the separate A, B, and C terms commonly used in the literature [4,5,2]. The operator expression (3.15) helps to understand the common origin of these terms. Moreover, our results are not restricted to the magnetic Faraday effect, but apply to any situation where the refractive index is modified by an axially symmetric perturbation. This allows us to apply these expressions also for the mechanical Faraday effect.

In the correction (3.25), each term has the frequency dependence of the derivative of a dispersion curve. This reflects its origin in the modified transition frequency. On the other hand, each term in (3.27) has the spectral characteristic of a dispersion curve, which might suggest that the physical origin of n_b is quite distinct from that of n_a. However, we can rewrite the second term in (3.27) by interchanging the summation indices e and e', so that combination with the first term yields the difference between two dispersion curves. When the difference between two unperturbed energy levels is smaller than the linewidth γ, the result can also be expressed as the derivative of dispersion curves. A similar treatment of the third and fourth terms yields the derivative of a dispersion curve weighted by a Boltzmann factor. Hence in the limit that two unperturbed energy levels approach each other, the term n_b does not go to zero, but becomes part of n_a and n_c.

Finally one notices that Doppler broadening can be included into our results, simply by replacing in (3.24)–(3.28) the frequency ω by $\omega-\mathbf{K}\cdot\mathbf{v}$, and taking the average over a Maxwell distribution.

IV. MECHANICAL FARADAY EFFECT IN A GAS

Now we turn to a discussion of the optical activity of gases induced by rotation of the vessel. When the gas is dilute, the gas particles spend most of their time in free flight. Therefore, as viewed from the laboratory frame, their internal states and the transition frequencies are unaffected by the rotation. The only effect is a modification of the internal state populations and of the distribution of velocities, resulting from a modified collision dynamics with the rotating wall. It is expected that the change in populations gives rise to a correction to the refractive indices of the type of n_c, whereas the rotational flow of the gas will give a kinematic contribution as in (2.13).

This expectation is confirmed if we apply the formalism of Sec. III. This formalism presupposes that the steady-state density matrix and the dynamical evolution of the system are described by a single Hamiltonian, which is only true in the rotating frame. After the gas particles have been fully accommodated to the rotating wall of the vessel, the translational state of the gas particles is given by a Boltzmann distribution with the perturbed Hamiltonian

$$H_{\text{tr}}=\frac{\mathbf{P}^2}{2M}-\boldsymbol{\Omega}\cdot\mathbf{L}_{\text{c.m.}}\ , \tag{4.1}$$

with $\mathbf{P}$ the momentum and $\mathbf{L}_{\text{c.m.}}$ the angular momentum of the center-of-mass motion of an atom. If we describe this center-of-mass motion of the gas particles classically, the distribution of the positions and the momenta of the centers of mass of the atoms in the rotating frame is given by the distribution function

$$f(\mathbf{P},\mathbf{R})=\frac{1}{W}\exp\left[-\beta\left[\frac{\mathbf{P}^2}{2M}-\boldsymbol{\Omega}\cdot(\mathbf{R}\times\mathbf{P})\right]\right]\ , \tag{4.2}$$

with W a normalization constant. The velocity distribution of the centers of mass of the gas particles is a Maxwellian, with zero average in the rotating frame. The centrifugal modification of the particle density is of second order in Ω, and can be neglected. Hence the density and the velocity distribution are rigidly corotating.

The effect of the frame rotation with angular velocity Ω on the dynamics of a gas particle is expressed by the perturbation Hamiltonian

$$H_1=-\Omega J_Z\ , \tag{4.3}$$

with $\mathbf{J}$ the total angular momentum of the internal state of the particle. With this perturbation the theory of Sec. III gives an expression for the correction n_1 to the refractive index *in the corotating frame.* The refractive indices $n_\pm$ as viewed in the laboratory frame are obtained by a transformation as applied by Player [12]. This yields the expressions

$$n_\pm(\omega)=n_0(\omega)\mp\Omega\frac{dn_0}{d\omega}\mp\frac{\Omega}{\omega}\left[n_0(\omega)-\frac{1}{n_0(\omega)}\right]+n_1(\omega,\mathbf{u}_\pm)\ . \tag{4.4}$$

Since the perturbation (4.3) commutes rigorously with the unperturbed Hamiltonian H_0 of a gas particle, the correction n_b vanishes. As one may expect from the above discussion, the term n_a exactly cancels the dispersive term in (4.4). This follows directly from the commutation rules for the Z component of the total angular momentum with the spherical components of a vector operator. This leads to the identity

$$[H_1,\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}]=-\hbar\Omega\mathbf{u}_+\cdot\boldsymbol{\mu}_{EG}\ . \tag{4.5}$$

Using this result in (3.25) leads to the simple relation

$$n_a(\omega,\mathbf{u}_+)=\Omega\frac{dn_0}{d\omega}\ . \tag{4.6}$$

Physically, this cancellation of the dispersive term shows that the internal state of the atoms does not follow the external rotation. With (2.4) we arrive at the expression for the rotation angle

$$\Theta_{\text{mech}}=\frac{\Omega Z}{c}\left[n_0(\omega)-\frac{1}{n_0(\omega)}\right]-\frac{\omega Z}{c}n_c(\omega,\mathbf{u}_+)\ . \tag{4.7}$$

By using (3.28), we find the expression

$$n_c(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_{e,g,M}|\langle e,M+1|\boldsymbol{\mu}_{EG}\cdot\mathbf{u}_+|g,M\rangle|^2\times\frac{-\beta\Omega Me^{-\beta E_g}}{Z_0}\,\text{Re}\frac{i}{\gamma-i(\omega-\omega_{eg})}\ . \tag{4.8}$$

Typically, the contribution from n_c to the angle Θ_{mech} is larger than the kinematic contribution by a factor of the order of $\hbar\omega/k_BT$. This result [(4.7) and (4.8)] is equally valid for an atomic or a molecular gas. Moreover, it remains valid in the presence of electron spin. Notice that the contribution from n_c was not obtained in the simplified picture of Sec. II. This is due to the fact that the Becquerel result (2.12) is equivalent to the magnetic term n_a, while the contribution from n_c is neglected. The terms n_c are nonzero only when the ground level has magnetic degeneracy.

It may be useful to briefly compare the results of this section with the work of Silverman [13]. He considered the setup where the complete system is mounted to a rotating frame, and he discussed the optical activity of a vapor of essentially free atoms, without any material wall. This means that as viewed from the inertial frame, the translational as well as the internal states of the atoms are unaffected by the rotation, so that the kinematic contribution and the term n_c vanish. On the other hand, since the source of linearly polarized light is corotating, the light beam as viewed in the inertial frame is composed of two circularly polarized beams, with frequencies $\omega \pm \Omega$. Hence, in the inertial frame the refractive indices $n_\pm$ are simply given by

$$n_\pm = n_0(\omega \pm \Omega) , \tag{4.9}$$

which leads with (2.4) to Silverman's result for the rotational angle

$$\Theta_{\text{mech}} = -\frac{\Omega Z}{c} \omega \frac{dn_0}{d\omega} . \tag{4.10}$$

One notices that this result may be viewed as arising from a rotational effect on the radiation field rather than on the atoms. In fact, there is an additional geometrical contribution arising from the fact that the two circular components with different frequencies also have different wavelengths.

V. MAGNETIC FARADAY EFFECT IN AN ATOMIC GAS

Since we are interested in the effects of electron spin, we shall apply the results of Sec. III to a fine-structure multiplet, without bothering about possible effects of the hyperfine structure [18,19]. For an atomic gas in a uniform magnetic field in the Z direction, the perturbation H_1 takes the form

$$H_1 = \frac{e}{2m} \mathbf{B} \cdot (\mathbf{L} + 2\mathbf{S}) = \frac{e}{2m} B (L_Z + 2S_Z) , \tag{5.1}$$

with $\mathbf{L}$ the total orbital angular momentum of the electrons, and $\mathbf{S}$ the total electron spin of the atom. This expression generalizes (2.6). The factor 2 in front of the spin arises from the different gyromagnetic ratio of the electron spin. In the present case of a magnetic field, the Hamiltonian H describes the dynamics of the electrons in the laboratory frame. Therefore the refractive indices $n_\pm$ in the laboratory frame are found from (3.15) or (3.29) when substituting (5.1) for H_1. Hence the rotation angle Θ is given by (2.4), if we substitute

$$n_\pm = n(\omega, \mathbf{u}_\pm) . \tag{5.2}$$

Using the symmetry relation (3.16) then gives

$$\Theta_{\text{mag}} = -\frac{\omega Z}{c} n_1(\omega, \mathbf{u}_+) , \tag{5.3}$$

where n_1 can be split into three parts according to (3.29).

A. Atomic singlet transition

In the case of a transition between singlet states in an atomic vapor, the spin contribution in (5.1) vanishes. The magnetic perturbation H_1 becomes identical to (4.3), with Ω replaced by $-eB/2m$. Therefore, the results of Sec. IV for the correction n_1 to the refractive index can be applied in the present case. Hence the term n_b vanishes, and n_a obeys an equality analogous to (4.6). In the present case of fully degenerate excited and ground levels, the density matrix σ_0 is isotropic and equivalent to the scalar $(2L_g+1)^{-1}$. The unperturbed refractive index (3.14) takes the well-known form

$$n_0(\omega) - 1 = \frac{N}{2\epsilon_0 \hbar} \frac{1}{3(2L_g+1)} |\langle E \| \mu \| G \rangle|^2 \frac{-\Delta}{\gamma^2 + \Delta^2} , \tag{5.4}$$

with $\Delta = \omega - \omega_{eg}$ the detuning of the light frequency from resonance. We have introduced the reduced dipole matrix element, which obeys the relation

$$\text{Tr}(\mathbf{u}^* \cdot \boldsymbol{\mu}_{GE} \boldsymbol{\mu}_{EG} \cdot \mathbf{u}) = \tfrac{1}{3} |\langle E \| \mu \| G \rangle|^2 , \tag{5.5}$$

independent of the polarization vector $\mathbf{u}$. The result for n_a is

$$\begin{aligned} n_a(\omega, \mathbf{u}_+) &= -\frac{eB}{2m} \frac{dn_0}{d\omega} \\ &= \frac{N}{2\epsilon_0 \hbar} \frac{1}{3(2L_g+1)} \\ &\quad \times |\langle E \| \mu \| G \rangle|^2 \frac{eB}{2m} \frac{\gamma^2 - \Delta^2}{(\gamma^2 + \Delta^2)^2} . \end{aligned} \tag{5.6}$$

Likewise, the term n_c is found by specializing (4.8) to the present case of a singlet transition, and substituting for Ω the appropriate value $-eB/2m$.

Comparison of these results with (4.7) shows that the complementarity relation (2.10) holds exactly for an atomic singlet transition. Over most of the frequency range, the ratio n_c/n_a is of the order of $\beta \hbar \Delta$, so that in the neighborhood of a resonance n_c is negligible in comparison with n_a. This means that in the complementarity relation (2.10), Θ_{mech} is much smaller than Θ_{mag}. Combination of (5.6) and (5.3) leads to the expression for the rotation angle

$$\Theta_{\text{mag}} = \frac{\omega Z}{c} \frac{eB}{2m} \frac{dn_0}{d\omega} , \tag{5.7}$$

which is equivalent to the Becquerel result (2.12). Note that the factor $eBL/2mc$ has the significance of the angle over which the precession occurs during the travel time of the light through the system.

B. Atomic multiplet transition

In the case of fine-structure multiplets in an atomic vapor, the spin is nonzero, and the perturbation (5.1) does not commute with the free-atom Hamiltonian H_0. The Faraday effect in the LS-coupling scheme has been treated by Rosenfeld [3] in the case of nonresonant radiation. We shall drop this restriction, and we express the results in terms of the general treatment of Sec. III. It is convenient to rewrite (5.1) in the form

$$H_1 = \frac{e}{2m} B (J_Z + S_Z) , \tag{5.8}$$

where $\mathbf{J} = \mathbf{L} + \mathbf{S}$ is the total electronic angular momentum. The term proportional to J_Z in (5.8) commutes with H_0, and only causes Zeeman shifts of the unperturbed levels. The additional spin term does not commute with H_0, and contributes both to H_d and to H_n. The diagonal part H_d is responsible for the anomalous Zeeman effect, and the nondiagonal part H_n mixes the states. Matrix elements of H_d and H_n can be evaluated for each specific case of values of L and S by standard Racah algebra.

We separate the raising part of the electric dipole in contributions from each transition by writing

$$\boldsymbol{\mu}_{EG}=\sum_e \sum_g \boldsymbol{\mu}_{eg} , \tag{5.9}$$

where e and g indicate the sublevels with quantum numbers J_e and J_g. A similar notation will be used for the lowering part and for the perturbation H_1. The unperturbed refractive index n_0 can be written as the sum of contributions from each component in the multiplet as

$$n_0(\omega)-1=\sum_g \sum_e n_{eg}(\omega) \tag{5.10}$$

with

$$n_{eg}(\omega)=\frac{N}{2\epsilon_0}\,\mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_{eg})}Z_0^{-1} \times \exp(-\beta E_g)\tfrac{1}{3}|\langle e\|\mu\|g\rangle|^2 . \tag{5.11}$$

The level spacings and the differences in transition frequencies are determined by the Landé rule for the spin-orbit coupling energy [20],

$$E_e=\tfrac{1}{2}A_e[J_e(J_e+1)-L_e(L_e+1)-S(S+1)] , \tag{5.12}$$

and likewise for the ground-state splitting.

Each of the three corrections (3.25), (3.27), and (3.28) can be expressed in terms of the factors

$$F(ee';g)=\mathrm{Tr}\,\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{ge}H_{1,ee'}\mathbf{u}_+\cdot\boldsymbol{\mu}_{e'g} \tag{5.13}$$

and

$$G(e;g'g)=\mathrm{Tr}\,\mathbf{u}_+^*\cdot\boldsymbol{\mu}_{g'e}\mathbf{u}_+\cdot\boldsymbol{\mu}_{eg}H_{1,gg'} , \tag{5.14}$$

where the traces are taken over the magnetic substates of the levels g or g'. These quantities are real, and they are symmetric in the sense that

$$F(ee';g)=F(e'e;g),\quad G(e;gg')=G(e;g'g) . \tag{5.15}$$

Explicit expressions are given in Appendix A. The three corrections to the refractive index are now

$$n_a(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_g \sum_e \mathrm{Re}\frac{1/\hbar^2}{[\gamma-i(\omega-\omega_{eg})]^2} \times Z_0^{-1}\exp(-\beta E_g) \times[F(ee;g)-G(e;gg)] , \tag{5.16}$$

$$n_b(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_g \sum_e \mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_{eg})}Z_0^{-1}\exp(-\beta E_g) \times\left[\sum_{e'\neq e}\frac{2F(ee';g)}{E_e-E_{e'}} + \sum_{g'\neq g}\frac{2G(e;gg')}{E_g-E_{g'}}\right] , \tag{5.17}$$

$$n_c(\omega,\mathbf{u}_+)=\frac{N}{2\epsilon_0}\sum_g \sum_e \mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_{eg})}Z_0^{-1} \times \exp(-\beta E_g)(-\beta)G(e;gg) . \tag{5.18}$$

In the usual situation that the temperature is sufficiently high, so that $k_B T$ is large compared with the level splittings, the distribution over the states is fully uniform. Then the Boltzmann factors in Eqs. (5.16) and (5.17) for n_a and n_b are determined by the statistical weights, and we can neglect the term n_c altogether. The magnitude of n_a is independent of the fine-structure level splittings, whereas the individual terms contributing to n_b are inversely proportional to these splittings. Hence, when the levels are well resolved, and the frequency is close to a transition frequency, the perturbation of the refractive index is well approximated by the contribution to n_a from that particular fine-structure transition. If Eqs. (A5) and (A6) are substituted into Eq. (5.16) for n_a, and the result is compared with (5.11), the relation

$$n_a(\omega,\mathbf{u}_+)=-\frac{eB}{2m}\frac{dn_{eg}}{d\omega}\{\tfrac{1}{4}g_e[2+J_e(J_e+1)-J_g(J_g+1)] +\tfrac{1}{4}g_g[2+J_g(J_g+1) -J_e(J_e+1)]\} \tag{5.19}$$

follows. The Landé factors g_e and g_g are given in Appendix A. These factors, which determine the splitting for the Zeeman components of each fine-structure component, also determine the effective frequency shift for each separate component. Therefore, when the frequency of the radiation is close to the fine-structure transition frequency between the levels e and g, the angle of rotation (2.4) takes the form

$$\Theta_{\mathrm{mag}}=\frac{\omega Z}{c}\frac{eB}{2m}\frac{dn_{eg}}{d\omega}\{\tfrac{1}{4}g_e[2+J_e(J_e+1)-J_g(J_g+1)] +\tfrac{1}{4}g_g[2+J_g(J_g+1)-J_e(J_e+1)]\} . \tag{5.20}$$

For spin zero, the factor in curly brackets is unity, and the Becquerel result is recovered.

In conclusion, a nonzero electron spin leads to a modification of all three terms contributing to the magnetic effect. For a single pair of sublevels, the largest contribution n_a is modified by a constant factor. On the other hand, the expressions for the mechanical effect are basically unaffected by spin. As a result, the complementarity relation (2.10) is invalid for an atomic transition between states with nonzero electron spin.

VI. ROTATIONAL BAND IN A MOLECULAR GAS

In this section we shall compare both Faraday effects in the rotational band spectrum of a diatomic gas. We consider the simplest possible case of a transition between two electronic singlet states, for a certain vibrational state of the ground and excited levels. This is another useful testing ground for the complementarity relation

(2.10). The molecular magnetic Faraday effect is reduced by the fact that the magnetic moment corresponding to the electronic angular momentum is partly averaged out by the molecular rotation. One may wonder whether the mechanical effect is accordingly augmented, so as to maintain the complementarity.

The total angular momentum of the molecules $\mathbf{J}$ is the sum of the electronic orbital angular momentum, and the contribution from the end-over-end rotation. In the Born-Oppenheimer approximation, the electronic states are characterized by the quantum number Λ for the component of the electronic angular momentum along the internuclear axis. The substates of the ground level are denoted as $|JM\Lambda\rangle$, and the excited substates as $|J'M'\Lambda'\rangle$. In Appendix B we give some relevant expressions for the dipole matrix elements between these substates as functions of the quantum numbers J, M, J' and M'. We shall consider the two complementary limiting cases of a single isolated line, and of a band of strongly overlapping lines.

A. Isolated rotational line

We introduce the function $f(\omega)$ as the fictitious refractive index corresponding to the electronic-vibrational transition that would arise if the rotational energies were fully negligible. In the case of an isolated rotational line, the unperturbed refractive index is then given by f at a frequency that is shifted by the rotational transition frequency, and multiplied by the appropriate Boltzmann population factor and branching ratio U. We obtain

$$n_0(\omega)-1=\frac{1}{Z_0}\exp(-\beta E_{J\Lambda})(2J+1)U(J,J';\Lambda,\Lambda') \times f\left[\omega-\frac{1}{\hbar}(E_{J\Lambda}-E_{J'\Lambda'})\right] \tag{6.1}$$

with Z_0 the purely rotational partition function. Expressions for the functions U and f are given in Appendix B, and the rotational contributions to the energy levels are [21]

$$\begin{aligned} E_{J\Lambda}&=\frac{\hbar^2}{2I}[J(J+1)-\Lambda^2]\ ,\\ E_{J'\Lambda'}&=\frac{\hbar^2}{2I'}[J'(J'+1)-\Lambda'^2] \end{aligned} \tag{6.2}$$

with I and I' the moment of inertia in the ground state and the excited state.

1. *Mechanical effect*

As shown in Sec. IV, the mechanical Faraday effect is determined by the term n_c, which is represented by (4.8). For an isolated rotational line the only remaining summation index is M, and since the dipole matrix elements in (4.8) are proportional to a $3j$ symbol, the summation is easily performed. The result can be put in terms of the unperturbed refractive index as

$$n_c(\omega,\mathbf{u}_+)=\beta\hbar\Omega W(J,J')[n_0(\omega)-1]\ , \tag{6.3}$$

where the factors W are given in Eqs. (B6) and (B7) of Appendix B. For a P line, the factor W is negative, indicating that the line gets weaker by the rotation. Conversely, an R line gets stronger. This may be understood by noting that the external rotation shifts population to higher M values. The corresponding contribution to the mechanical Faraday rotation angle Θ_{mech}, which is given in (4.7), is again larger than the kinematic contribution by a factor of the order of $\hbar\omega/k_BT$.

2. *Magnetic effect*

For a diatomic molecule in the state $|JM\Lambda\rangle$ in a magnetic field, the perturbation Hamiltonian is

$$H_1=\frac{e\hbar}{2m}B\Lambda\hat{\mathbf{Z}}\cdot\hat{\mathbf{z}} \tag{6.4}$$

with $\hat{\mathbf{Z}}$ the unit vector in the direction of the magnetic field and $\hat{\mathbf{z}}$ the unit vector along the internuclear axis. This perturbation is not simply proportional to the perturbation (4.3) induced by external rotation. The diagonal elements of H_1, which correspond to the Zeeman splittings of the lines, are equal to [22]

$$\langle JM\Lambda|H_1|JM\Lambda\rangle=\frac{e\hbar B}{2m}\frac{M\Lambda^2}{J(J+1)}\ . \tag{6.5}$$

When J is large compared with Λ, the Zeeman splitting is small. The Zeeman terms (6.5) determine the corrections n_a and n_c to the refractive index. If we substitute (6.5) into Eq. (3.25), and use expressions for the dipole matrix elements as specified by Eqs. (B2) and (B3), the summation over the quantum number M can be performed. After application of Eq. (6.1) for n_0, we arrive at a generalization of the Becquerel result,

$$n_a(\omega,\mathbf{u}_+)=-\frac{eB}{2m}\frac{dn_0}{d\omega}C(J,J';\Lambda,\Lambda')\ , \tag{6.6}$$

where for a P, Q, or R line the factors C are given by

$$\begin{aligned} C(J,J-1;\Lambda,\Lambda')&=\frac{-\Lambda'^2+\Lambda^2}{2J}\ ,\\ C(J,J;\Lambda,\Lambda')&=\frac{\Lambda'^2+\Lambda^2}{2J(J+1)}\ ,\\ C(J,J+1;\Lambda,\Lambda')&=\frac{\Lambda'^2-\Lambda^2}{2(J+1)}\ . \end{aligned} \tag{6.7}$$

The reduction factors C are smaller than 1, and decrease for increasing values of J. This reflects that the magnetic moment, which is oriented along the internuclear axis, is effectively reduced by the rotation of the molecule. Substituting (6.5) into (3.28), while using (6.1), leads to the result

$$n_c(\omega,\mathbf{u}_+)=-\beta\frac{e\hbar B}{2m}\frac{\Lambda^2}{J(J+1)}W(J,J')[n_0(\omega)-1]\ , \tag{6.8}$$

with the factors W given in (B7). In the usual case that $\hbar\gamma \ll k_BT$, the correction n_c is much smaller than n_a near resonance. Furthermore, the perturbation (6.4) has off-diagonal elements between states with different J

values, which give rise to a term n_b. However, for a well-isolated line, the linewidth is small compared with the line separation, and the correction n_b is negligible.

Hence, the Faraday rotation angle Θ_{mag} is given by Eq. (5.3), where n_1 is well approximated by n_a as given in (6.6). The decrease of the factors C for increasing values of J is not compensated by a corresponding increase of a dispersive term in the mechanical effect, as would be required to validate the complementarity derived in Sec. II. Likewise, the magnetic n_c term (6.8) does not exactly cancel the mechanical term (6.3) for a magnetic field obeying the Larmor condition (2.7). Hence, a magnetic field cannot compensate the Coriolis force, and the complementarity of the two Faraday effects does not hold near an isolated rotational resonance. This is due to the quantum-mechanical nature of molecular rotation. Only in the case that the rotational level structure is ineffective can we expect the result of Sec. II to apply.

B. Rotational band

In spectral regions where several rotational lines contribute to a single electronic-vibrational transition, the unperturbed refractive index can be written as

$$n_0(\omega)-1=\sum_J\sum_{J'}\frac{1}{Z_0}\exp(-\beta E_{J\Lambda})(2J+1)U(J,J';\Lambda,\Lambda')\times f\left[\omega-\frac{1}{\hbar}(E_{J\Lambda}-E_{J'\Lambda'})\right], \tag{6.9}$$

which is simply a summation over single-line contributions (6.1). The line profile f is assumed to vary only slightly over a rotational splitting. This is justified either for very large (homogeneous or inhomogeneous) linewidths, or for outside a rotational band. In this case the quantum nature of the rotation becomes unimportant.

1. Mechanical effect

The rotation angle of the mechanical effect is given by (4.7) in terms of the first-order correction n_c. This term can be expressed as

$$n_c(\omega,\mathbf{u}_+)=\sum_J\sum_{J'}\frac{1}{Z_0}\exp(-\beta E_{J\Lambda})(2J+1)U(J,J';\Lambda,\Lambda')\beta\hbar\Omega W(J,J')f\left[\omega-\frac{1}{\hbar}(E_{J\Lambda}-E_{J'\Lambda'})\right]. \tag{6.10}$$

The expressions (6.9) and (6.10) take a simple form after expanding the function f to first order, provided that the moments of inertia I and I' are the same. Introducing the average change of rotational energy at excitation of a ground state with angular momentum J,

$$\langle\Delta E\rangle=\sum_{J'}U(J,J';\Lambda,\Lambda')(E_{J\Lambda}-E_{J'\Lambda'}), \tag{6.11}$$

gives the result

$$\begin{aligned}\langle\Delta E\rangle&=\frac{\hbar^2}{2I}\quad\text{for }\Lambda'=\Lambda\pm1\\ \langle\Delta E\rangle&=\frac{\hbar^2}{I}\quad\text{for }\Lambda'=\Lambda\,.\end{aligned} \tag{6.12}$$

Therefore up to first order Eq. (6.9) gives the identity

$$\begin{aligned}n_0(\omega)-1&=f\left[\omega-\frac{\hbar}{2I}\right]\quad\text{for }\Lambda'=\Lambda\pm1\\ n_0(\omega)-1&=f\left[\omega-\frac{\hbar}{I}\right]\quad\text{for }\Lambda'=\Lambda\,.\end{aligned} \tag{6.13}$$

A similar expansion of (6.10) to first order, and substitution of the explicit values of the $3j$ symbols in U, gives

$$\begin{aligned}n_c(\omega,\mathbf{u}_+)&=\pm\tfrac{1}{2}\beta\hbar\Omega\Lambda f(\omega)\\ &\quad+\frac{\Omega}{2}\frac{dn_0}{d\omega}\frac{\beta}{Z_0}\frac{dZ_0}{d\beta}\quad\text{for }\Lambda'=\Lambda\pm1\\ n_c(\omega,\mathbf{u}_+)&=\Omega\frac{dn_0}{d\omega}\frac{\beta}{Z_0}\frac{dZ_0}{d\beta}\quad\text{for }\Lambda'=\Lambda\,.\end{aligned} \tag{6.14}$$

The only approximation made so far is the expansion to first order of the profile around the average frequency. In particular, these results are valid for arbitrary values of the temperature. Notice that the average rotational energy is equal to

$$\langle E\rangle=-\frac{1}{Z_0}\frac{dZ_0}{d\beta}\,. \tag{6.15}$$

In fact, the small rotational frequency shifts in (6.13) may be ignored, so that we can simply equate f to n_0-1.

When the temperature is sufficiently high, the rotation can be described classically, and $\langle E\rangle=\beta^{-1}$. Then (6.14) can be replaced by the fully classical result

$$\begin{aligned}n_c(\omega,\mathbf{u}_+)&=\pm\tfrac{1}{2}\beta\hbar\Omega\Lambda[n_0(\omega)-1]\\ &\quad-\frac{\Omega}{2}\frac{dn_0}{d\omega}\quad\text{for }\Lambda'=\Lambda\pm1\\ n_c(\omega,\mathbf{u}_+)&=-\Omega\frac{dn_0}{d\omega}\quad\text{for }\Lambda'=\Lambda\,.\end{aligned} \tag{6.16}$$

Finally, substitution of (6.16) in (4.10) gives for the Faraday rotation angle the result

$$\begin{aligned}\Theta_{\text{mech}}&=\frac{\Omega Z}{c}\left[n_0(\omega)-\frac{1}{n_0(\omega)}+\tfrac{1}{2}\omega\frac{dn_0}{d\omega}\right.\\ &\qquad\left.\mp\tfrac{1}{2}\beta\hbar\omega\Lambda[n_0(\omega)-1]\right]\quad\text{for }\Lambda'=\Lambda\pm1\\ \Theta_{\text{mech}}&=\frac{\Omega Z}{c}\left[n_0(\omega)-\frac{1}{n_0(\omega)}+\omega\frac{dn_0}{d\omega}\right]\quad\text{for }\Lambda'=\Lambda\,.\end{aligned} \tag{6.17}$$

These results are obtained by adopting three assumptions, which imply a fully classical picture of the molecular rotation. First, the line profiles are assumed to vary only linearly over rotational level splittings. Second, the moments of inertia in the ground level and the excited level are taken the same. Finally, the classical expression for the average rotational energy is assumed to hold.

In the case that $\Lambda'=\Lambda\pm1$, the term proportional to n_0-1 in (6.17) results from the change in the transition strength due to the modified populations. The frequency derivatives result from the decreased strength of the P transitions and the increased strength of the R transitions. In the case that $\Lambda'=\Lambda$, the total transition strength is evenly distributed over the P and R bands, whereas the Q band is absent. For $\Lambda'=\Lambda\pm1$, half of the transition strength goes into the Q band, and only the other half remains for the P and the R bands. This explains the different factor in front of the derivatives in (6.17). For $\Lambda'=\Lambda$ the mechanical rotation angle is identical to $\Theta_{\rm rig}$, Eq. (2.9), corresponding to rigid rotation. Hence, the macroscopic rotation is completely transferred to the electronic wave functions. For $\Lambda'=\Lambda\pm1$ this transfer is only 50%. The physical reason for this difference is that the radiative transition in the case that $\Lambda'=\Lambda\pm1$ occurs preferably when the internuclear axis is parallel or antiparallel to the propagation direction, where the circular polarization vector imposes the right selection rule. But when the internuclear axis coincides with the rotation axis, the slip is maximal. For $\Lambda'=\Lambda$, the transition selects the component of the polarization vector that is parallel to the internuclear axis. Hence the transition probability is maximal when the internuclear axis falls in the XY plane, where in the Born-Oppenheimer approximation the rotary drag is complete.

2. *Magnetic effect*

At first sight, the magnetic Faraday effect is more complex than the mechanical effect, since the magnetic perturbation H_1 as given in (6.4) has nondiagonal elements in the rotational quantum number J. However, the result greatly simplifies if we ignore the rotational contribution to the transition frequencies. Then one may start from Eq. (3.15), where the Liouville operator $\mathcal{L}_0$ can simply be replaced by the single eigenvalue ω_0. Both the transition dipoles and the perturbation (6.4) take the form of Wigner rotation matrices, and the total correction to the refractive index is

$$n_1(\omega,\mathbf{u}_+)=\sum_{J'}\frac{1}{Z_0}\exp(-\beta E_{J\Lambda})\frac{eB}{2m}A(J,J';\Lambda,\Lambda')\times\left[-(\Lambda'-\Lambda)\frac{df}{d\omega}-\beta\hbar\Lambda f(\omega)\right], \quad (6.18)$$

where the function A, defined in Eq. (B8) in Appendix B, attains the simple values given in (B9). Hence we arrive at the final result

$$n_1(\omega,\mathbf{u}_+)=-\frac{1}{2}\frac{eB}{2m}\left[(\Lambda'-\Lambda)^2\frac{dn_0}{d\omega}+\beta\hbar(\Lambda'-\Lambda)\Lambda[n_0(\omega)-1]\right]. \quad (6.19)$$

The term proportional to n_0-1 corresponds to n_c, and it changes sign with $\Lambda'-\Lambda$. The term proportional to the derivative is the same for $\Lambda'-\Lambda=\pm1$, and arises from n_a+n_b. It is remarkable that Eq. (6.16), which has a rather similar structure, is purely of the type n_c. In order to obtain the Faraday rotation angle, we substitute (6.19) into (5.3), with the result

$$\Theta_{\rm mag}=\begin{cases}\dfrac{1}{2}\dfrac{eB}{2m}\dfrac{Z}{c}\left[\omega\dfrac{dn_0}{d\omega}\pm\beta\hbar\omega\Lambda[n_0(\omega)-1]\right] & \text{for } \Lambda'=\Lambda\pm1\\ 0 \text{ for } \Lambda'=\Lambda\,. \end{cases} \quad (6.20)$$

The complete vanishing of the Faraday effect for $\Lambda'=\Lambda$ is a feature of the Born-Oppenheimer approximation.

Combining Eqs. (6.20) and (6.17), it is easy to check that their sum exactly obeys the complementarity relation (2.10), for all possible values of $\Lambda'-\Lambda$. This complementarity is only valid under the condition that the rotation can be described in a fully classical fashion.

VII. CONCLUSIONS

We have compared the Faraday effect in gases with its mechanical analog. This comparison is relevant to the fundamental question under what conditions it is possible to rotate an atom [14]. From simple arguments one may conclude that the two effects are complementary in the sense of Eq. (2.10). This relation states that for a magnetic field B which is related to the angular velocity Ω by the Larmor condition $B=2m\Omega/e$, the sum of the two Faraday rotation angles is equal to the mechanical Faraday effect for a sample in fully rigid rotation. This latter effect contains two contributions: a kinematic term, resulting from the rotary motion of the centers of mass of the gas particles, and a dispersive term, arising from the fact that circularly polarized light with frequency ω propagating along the rotation axis is experienced by a rotating sample at the shifted frequency $\omega\pm\Omega$. Near resonance, the dispersive term is larger than the kinematic term by a factor of the order of ω/γ, with γ the total linewidth. Since the sum of the two Faraday effects is determined by only the refractive index of the sample at rest with zero magnetic field, the validity of (2.10) would mean that both effects contain equivalent information on the dynamics of the system. It has been our purpose in the present paper to investigate under which conditions this conclusion is valid. The analysis is based on a unified description of the perturbed refractive index of the vapor.

The case of a dilute atomic gas is discussed in Secs. IV and V. We show that the mechanical effect contains two terms. The first one is the purely kinematic one, arising from the fact that the velocity distribution follows the external rotation. The second term corresponds to the perturbation n_c of the refractive index. This perturbation is analogous to a paramagnetic term, and results from the modification of the populations of the substates (if any) of the ground level. This latter contribution to the Faraday rotation angle is larger than the kinematic effect by a factor $\hbar\omega/k_BT$, provided that the lower level is degenerate. The magnetic effect is given by the classical Becquerel result and a small paramagnetic correction resulting from the modified ground-state populations. This correction is smaller than the Becquerel term by a factor of the order of $\hbar\gamma/k_BT$. For a magnetic field B related to the angular velocity by the Larmor relation (2.7), the paramagnetic contributions are each other's opposite. But the magnetic (Becquerel) effect is much larger than the kinematic term in the mechanical effect, by a factor of the order of ω/γ. We find that the complementarity relation of the two effects holds exactly. For atoms with nonzero electron spin, the complementarity no longer holds. The mechanical effect remains basically the same (i.e., small), whereas the magnetic effect remains large, but becomes quite different.

In Sec. VI the case of a diatomic gas with zero electron spin is considered. It is demonstrated that in the neighborhood of an isolated rotational line the complementarity relation (2.10) breaks down. Apparently, the derivation of this relation is not valid, due to the quantum character of the molecular rotation. Only in the limit that the rotational structure in the spectrum is negligible does the complementarity hold. This is correct for overlapping linewidths of the individual rotational lines, or beyond the bandhead.

These results indicate that the complementarity of the two Faraday effects, which is a direct result from Larmor's theorem, requires that the electrons are subject to a classical external rotation. Nonclassical aspects, such as rotational quantum levels and electron spin, spoil the complementarity.

In this paper we have assumed that the dynamics of the electrons in a gas particle (atom or molecule) is described by the Hamiltonian of the free gas particles. Perturbations by neighboring particles are accounted for by a possible broadening of the homogeneous linewidth γ. This corresponds to the impact approximation of the spectral lines, which is justified when the time between collisions is large compared with the duration of a collision. As indicated above, in this case the mechanical Faraday effect consists only of the kinematic and paramagnetic terms, which are much smaller than the Becquerel term of the magnetic Faraday effect. When the gas is sufficiently dense, or when the absorbing particles are embedded in a sufficiently dense buffer gas, so that multiple collisions are not negligible, inhomogeneous or quasistatic line broadening becomes dominant, and the present results are not valid [14]. When an atomic transition is perturbed by the presence of another atom, the Zeeman degeneracy of the angular momentum substates is lifted. When, moreover, the internuclear axis of the collision complex does not coincide with the rotation axis, the Coriolis term (4.3) will perturb the energy eigenstates of the collision complex. This means that the mechanical Faraday effect can be much larger than predicted by the present description. Conversely, the magnetic Faraday effect will be diminished in a complementary way. When the quantum structures of the rotation and spin effects are negligible, the mechanical and magnetic effects contain basically the same physics.

Although the complementarity relation indicates a close connection between the two Faraday effects, their practical measurability is quite different. Let us first consider an atomic gas. Even for moderate magnetic fields, the Larmor precession frequency $eB/2m$, which amounts to $2\pi\times1.4$ MHz for $B=1$ G, is much larger than practical angular velocities of mechanical rotation, which are of the order of $\Omega=2\pi\times100$ Hz. Thus, in the mechanical case, assuming rigid rotation, small rotation angles are expected. At present, the best polarimetric sensitivity of the rotation angle amounts to about $\simeq10^{-8}$ rad [23]. Of course, one may try to enlarge the rotation angle by using a longer cell. However, the length of the cell is obviously limited by absorption. When the length is chosen equal to the absorption length, the rotation angle is independent of the strength of a line [14]. A rough estimate of the linewidths γ for an alkali-metal vapor immersed in a very dense buffer gas, where presumably the rotation is rigid, indicates that a direct observation of the dispersive contribution to the mechanical effect is out of reach by one or two orders of magnitude [14]. However, progress in polarimetry may close this gap in the near future [13]. Far from resonance, only the kinematic and "paramagnetic" contributions remain, which are much smaller anyway. In the present paper we have stressed, however, that a measurement of the magnetic Faraday effect contains equivalent information (at least for zero spin).

For a molecular gas, the mechanical Faraday effect will be even smaller than for an atomic gas, due to the much smaller oscillator strength and ground-level population (if we are close to an individual line), or due to the large effective detuning (if we are beyond a bandhead). Also the magnetic Faraday effect near a molecular line is relatively small, due to the flywheel action of the molecular rotation, which diminishes the net magnetic moment [Eq. (6.5)].

Experimentally, therefore, the most promising route for studying the issue of rigid electronic rotation versus electronic slip seems to be a study of the magnetic Faraday effect of a high-density atomic gas. Experiments along these lines are presently in progress.

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APPENDIX A

Explicit expressions for the quantities F and G defined in (5.13) and (5.14) in the LS-coupling scheme, obtained by standard Racah algebra [20,24], contain the reduced matrix elements and $6j$ symbols

$$F(ee';g)=\frac{e\hbar B}{2m}\langle e'\|\mu\|g\rangle\langle e\|\mu\|g\rangle^*(-1)^{J_e+J_g}\begin{Bmatrix}1 & 1 & 1\\ J_g & J_{e'} & J_e\end{Bmatrix}\left[\frac{J_e(J_e+1)(2J_e+1)}{6}\right]^{1/2}$$
$$\times\left[\delta_{ee'}+(-1)^{S+J_{e'}+1+L_e}\begin{Bmatrix}S & S & 1\\ J_{e'} & J_e & L_e\end{Bmatrix}\left[\frac{S(S+1)(2S+1)(2J_{e'}+1)}{J_e(J_e+1)}\right]^{1/2}\right] \tag{A1}$$

and

$$G(e;gg')=-\frac{e\hbar B}{2m}\langle e\|\mu\|g\rangle\langle e\|\mu\|g'\rangle^*(-1)^{J_{g'}+J_e}\begin{Bmatrix}1 & 1 & 1\\ J_{g'} & J_g & J_e\end{Bmatrix}\left[\frac{J_g(J_g+1)(2J_g+1)}{6}\right]^{1/2}$$
$$\times\left[\delta_{gg'}+(-1)^{S+J_{g'}+1+L_g}\begin{Bmatrix}S & S & 1\\ J_{g'} & J_g & L_g\end{Bmatrix}\left[\frac{S(S+1)(2S+1)(2J_{g'}+1)}{J_g(J_g+1)}\right]^{1/2}\right]. \tag{A2}$$

In the LS scheme, the electric dipole commutes with the spin, and the reduced matrix elements for the J sublevels are related to a single matrix element for the multiplet, according to the relation [20]

$$\langle e\|\mu\|g\rangle=(-1)^{L_e+J_g+1+S}\sqrt{(2J_e+1)(2J_g+1)}\begin{Bmatrix}L_g & L_e & 1\\ J_e & J_g & S\end{Bmatrix}\langle E\|\mu\|G\rangle\ . \tag{A3}$$

Therefore, the terms F and G are all real.

In the diagonal case that $e=e'$ or $g=g'$, the factors in square brackets in (A1) and (A2) are just the Landé factors g_e and g_g of the levels, which are given by [20]

$$g=1+\frac{S(S+1)+J(J+1)-L(L+1)}{2J(J+1)}\ , \tag{A4}$$

where J takes the value of J_e or J_g, and where L stands for L_e or L_g. The diagonal values of the functions take the form

$$F(ee;g)=\tfrac{1}{12}|\langle e\|\mu\|g\rangle|^2\frac{e\hbar B}{2m}[2+J_e(J_e+1)-J_g(J_g+1)]g_e\ , \tag{A5}$$

$$G(e;gg)=-\tfrac{1}{12}|\langle e\|\mu\|g\rangle|^2\frac{e\hbar B}{2m}[2+J_g(J_g+1)-J_e(J_e+1)]g_g\ . \tag{A6}$$

APPENDIX B

The state of a diatomic molecule with zero electron spin, with quantum numbers J and M for the total angular momentum, and Λ for the component of the electronic angular momentum along the internuclear axis is denoted as $|JM\Lambda\rangle$. The normalized wave functions for the orientation of the internuclear axis indicated by the polar angles ϕ and θ are expressed in terms of the Wigner rotation matrices as [21]

$$\langle\phi,\theta|JM\Lambda\rangle=\left[\frac{2J+1}{8\pi^2}\right]^{1/2}\mathcal{D}^J_{\Lambda M}(\phi,\theta,0) \tag{B1}$$

in the notation convention of Edmonds [25]. The transition dipole between two molecular states can also be expressed in terms of a rotation matrix

$$\langle J'M'\Lambda'|\boldsymbol{\mu}_{EG}\cdot\mathbf{u}_+|JM\Lambda\rangle=D\langle J'M'\Lambda'|\mathcal{D}^1_{\Lambda'-\Lambda,1}(\phi,\theta,0)|JM\Lambda\rangle\ , \tag{B2}$$

where D is the dipole matrix element for the electronic and vibrational transition. This expression can be evaluated by using standard expressions for integrals of products of Wigner matrices, in the form

$$\langle J'M'\Lambda'|\mathcal{D}^j_{\lambda m}(\phi,\theta,0)|JM\Lambda\rangle=\sqrt{(2J+1)(2J'+1)}\times\begin{bmatrix}j & J & J'\\ \lambda & \Lambda & -\Lambda'\end{bmatrix}\begin{bmatrix}j & J & J'\\ m & M & -M'\end{bmatrix}. \tag{B3}$$

It is convenient to express the refractive index in terms of the function

$$f(\omega)=\tfrac{1}{3}|D|^2\frac{N}{2\epsilon_0}\,\mathrm{Re}\frac{i/\hbar}{\gamma-i(\omega-\omega_0)}\ , \tag{B4}$$

which is the contribution to the refractive index of a gas of two-state particles with transition frequency ω_0, and reduced dipole D. If we take for ω_0 the electronic plus vibrational contribution to the transition frequency, the unperturbed refractive index near an isolated rotational line is given by (6.1), with the branching ratios

$$U(J,J';\Lambda,\Lambda')=(2J'+1)\begin{bmatrix}1 & J & J'\\ \Lambda'-\Lambda & \Lambda & -\Lambda'\end{bmatrix}^2 \tag{B5}$$

These ratios add up to 1 after summation over J', and determine the relative strengths of the transitions with $J'=J-1$, $J'=J$, and $J'=J+1$, contributing to the P, Q, and R branch of the spectrum. We also need the factors

$$W(J,J')=3\sum_{M,M'}M\begin{bmatrix}1 & J & J'\\ 1 & M & -M'\end{bmatrix}^2 . \tag{B6}$$

Their explicit values are for a P line, a Q line, and an R line:

$$W(J,J-1)=-\frac{J+1}{2},\quad W(J,J)=-\tfrac{1}{2}, \tag{B6}$$
$$=W(J,J+1)=\frac{J}{2}\ . \tag{B7}$$

The magnetically perturbed refractive index in a rotational band is expressed in terms of the functions A, which are defined by

$$A(J,J';\Lambda,\Lambda')=3\sum_{M}\langle JM\Lambda|\mathcal{D}^{1*}_{\Lambda'-\Lambda,1}(\phi,\theta,0)\mathcal{D}^{1}_{00}(\phi,\theta,0)\mathcal{D}^{1}_{\Lambda'-\Lambda,1}(\phi,\theta,0)|JM\Lambda\rangle\ . \tag{B8}$$

After using identities for products of Wigner functions [25], one may show that

$$A(J,J';\Lambda,\Lambda')=\frac{2J+1}{2}(\Lambda'-\Lambda)\ . \tag{B9}$$

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