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Recoil-induced optical Faraday rotation

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It is shown that, as a result of recoil during spontaneous emission, optical activity in a vapor can be induced under the action of resonant pump and probe fields. For field geometries that exclude other mechanisms for rotation of the probe field's plane of polarization, the recoil accompanying spontaneous emission provides the needed coupling between the spatial and internal atomic degrees of freedom to produce an optically active medium. A transition between ground and excited states, each having angular momentum $J=\frac{1}{2}$, is analyzed in detail. The effect can be interpreted as the result of pump field scattering off a spatial polarization grating of ground-state Zeeman coherence.

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I. INTRODUCTION

In this article we show that the recoil an atom undergoes during spontaneous emission can lead to a rotation of the plane of polarization of a probe field when both the probe field and a pump field are incident on an initially isotropic vapor. We refer to this process as a recoil-induced optical Faraday rotation (RIOFR). It has already been pointed out that atomic recoil associated with stimulated processes [1] can lead to recoil-induced resonances (RIR's) in the pump-probe spectroscopy of a sub-Doppler [2] or subrecoil [3] cooled gas. When the pump and probe fields are detuned from the atomic transition frequency by an amount greater than the spontaneous decay rate, the RIR's can be understood as Raman-type two-quantum transitions between atomic states having different center-of-mass momenta [2]. When the fields are tuned close to resonance, the recoil associated with spontaneous emission [4] also contributes to the RIR signal [2] and can lead to qualitatively new effects in pump-probe spectroscopy. One such effect, RIOFR, is the subject of this contribution.

Consider the response of an atomic vapor to a pump and probe field. The total electric field vector is given by

$$\mathbf{E}(\mathbf{r},t)=\tfrac{1}{2}[E\mathbf{e}\exp(i\mathbf{k}\cdot\mathbf{r}-i\Omega t)+E'\mathbf{e}'\exp[(i\mathbf{k}'\cdot\mathbf{r}-i\Omega' t)] + \text{c. c.}, \qquad (1)$$

where $(E,\mathbf{e},\mathbf{k},\Omega)$ and $(E',\mathbf{e}',\mathbf{k}',\Omega')$ are the amplitude, polarization vector, wave vector, and frequency of the pump and probe fields, respectively. Optical Faraday rotation (OFR) [5] of the plane of polarization of the probe field can occur even without the inclusion of recoil when the pump field's polarization vector $\mathbf{e}$ satisfies $\mathbf{e}\cdot(\mathbf{k}'\times\mathbf{e}')\neq 0$ and $\mathbf{e}\cdot\mathbf{e}'\neq 0$. If, on the other hand, the probe field is polarized along $\hat{\mathbf{x}}$ and propagates along $\hat{\mathbf{z}}$ (Fig. 1) and the pump field propagates along $\hat{\mathbf{y}}$ with its polarization vector $\mathbf{e}$ in the $(\hat{\mathbf{x}},\hat{\mathbf{z}})$ plane, there is no OFR since $\mathbf{e}\cdot(\mathbf{k}'\times\mathbf{e}')=0$.

Nevertheless, the chosen geometry is asymmetric with respect to reflection relative to the probe field's plane of polarization, i.e., the sign of the pump field wave vector $\mathbf{k}$ changes under reflection about this plane. A dependence of the spectrum on the fields' propagation directions arises from the Doppler frequency shift $\mathbf{k}\cdot\mathbf{p}/m$, where $\mathbf{p}$ and m are the atomic momentum and mass. One can exploit this asymmetry to produce an optically active medium, if the spatial degrees of freedom (atomic momentum $\mathbf{p}$) are coupled to the internal degrees of freedom (magnetic quantum numbers of the involved atomic sublevels). Recoil associated with spontaneous emission provides this coupling. In this paper, we consider RIOFR on a $J=\frac{1}{2}$ to $J=\frac{1}{2}$ transition. The more general case of arbitrary angular momenta will be treated in a future paper.

It should be noted that a recoil-induced optical Faraday rotation has been considered under somewhat different conditions. Kazantsev *et al.* [6] showed that optical activity in an atomic beam could result when a linearly polarized field interacts with the atomic beam, provided recoil is included. The spontaneously emitted radiation together with the incident field couples the axial symmetry of the atomic beam to the internal states of the atom. On the other hand, in the example we discuss below, the spontaneous radiation couples an excited-state Zeeman coherence created by the pump and probe fields to a different component of the ground-state Zeeman coherence. Moreover, the angle of Faraday rotation varies as the pump field intensity for our pump-probe geometry, whereas it varies as the field intensity squared for the single field-atomic beam geometry considered in [6].

II. RIOFR ON A $\frac{1}{2}\rightarrow\frac{1}{2}$ TRANSITION

To illustrate RIOFR, we assume that pump and probe fields drive transitions between states each having total angular momentum $J=\frac{1}{2}$. In the absence of any applied fields, atomic density-matrix elements are given by

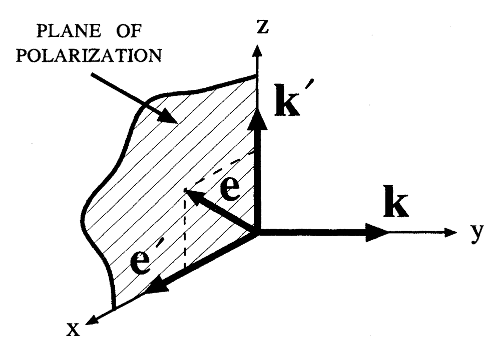

FIG. 1. Field geometry. The vectors $(\mathbf{k},\mathbf{e})$ and $(\mathbf{k}',\mathbf{e}')$ refer to the pump and probe fields, respectively. For this geometry, there is no optical Faraday rotation in the absence of recoil.

$$\langle \mathbf{p},m|\rho(i,j)|\mathbf{p}',m'\rangle = \tfrac{1}{2}\delta_{mm'}\delta_{i1}\delta_{j1}[(2\pi\hbar)^3/V]\delta(\mathbf{p}-\mathbf{p}')W(\mathbf{p}), \quad (2)$$

where ($i,j=1$ or 2) label the ground (1) and excited (2) states; m, $m'=\pm\frac{1}{2}$ are magnetic quantum numbers, $\mathbf{p}$ and $\mathbf{p}'$ are center-of-mass momenta, $W(\mathbf{p})$ is the atomic momentum distribution function, and V is the quantization volume. For a $\frac{1}{2}\rightarrow\frac{1}{2}$ transition, there are 16 density-matrix elements that can be divided into four 2×2 matrices $\rho(i,j)$ having matrix elements $\rho(i,j)_{m,m'}=\rho_{i,m;j,m'}$ ($i,j=1$ or 2). The matrices $\rho(1,1)$ and $\rho(2,2)$ characterize the ground- and excited-state populations and Zeeman coherences, while $\rho(1,2)$ and $\rho(2,1)$ characterize the ground-excited-state electronic coherence. The $\rho(i,j)$ can be represented in terms of "spin" by

$$\rho(i,j)=\tfrac{1}{2}\mathrm{Tr}[\rho(i,j)]+\mathbf{s}_{ij}\cdot\boldsymbol{\sigma}, \quad (3)$$

where $\boldsymbol{\sigma}=(\sigma_x,\sigma_y,\sigma_z)$ is composed of the Pauli spin matrices. Note that

$$(s_{ij})_x=\tfrac{1}{2}(\rho_{i,1/2;j,-1/2}+\rho_{i,-1/2;j,1/2}),$$

$$(s_{ij})_y=i/2(\rho_{i,1/2;j,-1/2}-\rho_{i,-1/2;j,1/2}),$$

$$(s_{ij})_z=\tfrac{1}{2}(\rho_{i,1/2;j,1/2}-\rho_{i,-1/2;j,-1/2}).$$

Of the many terms contributing to the excited-state density matrix in second-order perturbation theory, the only term that is relevant for the RIOFR (for our given geometry) is one that involves the combined action of the pump and probe fields. Explicitly one finds that, to order $E'E^*$, the excited-state spin has matrix elements

$$\langle \mathbf{p}+\hbar\mathbf{k}'|\mathbf{s}_{22}|\mathbf{p}'+\hbar\mathbf{k}\rangle = i[\chi'\chi^*/4(\gamma^2+\Delta^2)](\mathbf{e}'\times\mathbf{e}) \times\exp(-i\delta t)[(2\pi\hbar)^3/V]W(\mathbf{p})\delta(\mathbf{p}-\mathbf{p}'), \quad (4)$$

where $\chi=\mu E/2\sqrt{3}\hbar$ and $\chi'=\mu E'/2\sqrt{3}\hbar$ are Rabi frequencies of the pump and probe fields, respectively, μ is a reduced matrix element of the dipole moment operator between states 2 and 1, $\gamma=\Gamma/2$, Γ is the excited-state decay rate, $\Delta=\Omega-\omega$, ω is the transition frequency, and $\delta=\Omega'-\Omega$. We assume that $ku\ll\Gamma$ (u is the most probable atomic velocity) [7] and $|\delta|\ll|\Delta|$.

Owing to spontaneous emission, the excited-state spin serves as a driving term for the ground-state spin via the evolution equation

$$[\partial/\partial t+\gamma_t+i\omega(\mathbf{p},\mathbf{p}')]\langle\mathbf{p}|\mathbf{s}_{11}|\mathbf{p}'\rangle = -\Gamma\int\frac{d\mathbf{n}}{4\pi}\mathbf{n}(\mathbf{n}\cdot\langle\mathbf{p}+\hbar\mathbf{q}|\mathbf{s}_{22}|\mathbf{p}'+\hbar\mathbf{q}\rangle), \quad (5)$$

where γ_t is some effective decay rate of the ground state ($\gamma_t\ll\Gamma$), $\omega(\mathbf{p},\mathbf{p}')=(\mathbf{p}^2-\mathbf{p}'^2)/2m\hbar$ is a frequency of transition between ground states having momenta $\mathbf{p}$ and $\mathbf{p}'$, $\mathbf{n}=\mathbf{q}/q$, and $\mathbf{q}$ is the propagation vector of the spontaneously emitted photon. The steady-state solution of Eqs. (4) and (5) is

$$\langle\mathbf{p}+\hbar\mathbf{k}'|\mathbf{s}_{11}|\mathbf{p}'+\hbar\mathbf{k}\rangle=-i\Gamma[\chi'\chi^*/4(\gamma^2+\Delta^2)]\int\frac{d\mathbf{n}}{4\pi}\mathbf{n}[\mathbf{n}\cdot(\mathbf{e}'\times\mathbf{e})]\{\gamma_t-i[\delta-\omega(\mathbf{p}+\hbar(\mathbf{k}'-\mathbf{q}),\mathbf{p}'+\hbar(\mathbf{k}-\mathbf{q}))]\}^{-1} \times\exp(-i\delta t)[(2\pi\hbar)^3/V]W(\mathbf{p})\delta(\mathbf{p}-\mathbf{p}'). \quad (6)$$

One can substitute Eq. (6) into an expression for the matrix element of electronic coherence "spin,"

$$\langle\mathbf{p}|\mathbf{s}_{21}|\mathbf{p}'\rangle=-\chi\exp(-i\Delta t)\langle\mathbf{p}-\hbar\mathbf{k}|\mathbf{e}\times\mathbf{s}_{11}|\mathbf{p}'\rangle/\sqrt{2}(\gamma-i\Delta) \quad (7)$$

and use this expression to obtain the positive frequency component of the medium's polarization via

$$\mathbf{P}_+=\sqrt{2/3}\mu^*\exp(-i\omega t)[NV/(2\pi\hbar)^3]\int d\mathbf{p}\,d\mathbf{p}'\exp[i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}/\hbar]\langle\mathbf{p}|\mathbf{s}_{21}|\mathbf{p}'\rangle, \quad (8)$$

where N is the density of the vapor. One arrives at

$$\mathbf{P}_+=-N\mu^*[\chi/\sqrt{3}(\gamma-i\Delta)][V/(2\pi\hbar)^3]\int d\mathbf{p}\,d\mathbf{p}'\exp[-i\Omega t+i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}/\hbar]\langle\mathbf{p}-\hbar\mathbf{k}|\mathbf{e}\times\mathbf{s}_{11}|\mathbf{p}'\rangle. \quad (9)$$

This contribution to the polarization acts as a driving term in Maxwell's equation for the probe fields, since it varies as $\exp(-i\Omega' t+i\mathbf{k}'\cdot\mathbf{r})$. All other contributions to the polarization (from excited-state density-matrix elements and the homogeneous term $\mathrm{Tr}[\rho(1,1)]$ of the ground-state density matrix) have been excluded since they do not lead to OFR.

One finds that the probe field amplitude exiting the sample is given by

$$\mathbf{E}'_{\mathrm{out}}=E'(\hat{\mathbf{x}}+\epsilon\hat{\mathbf{y}}), \quad (10a)$$

where

$$\epsilon=-(\pi^{3/2}/6\sqrt{2})(N|\mu|^2 l/\hbar u)[|\chi|^2\Gamma/(\gamma^2+\Delta^2)(\gamma-i\Delta)]\int\frac{d\mathbf{n}}{4\pi}\{(\mathbf{e}\times\mathbf{n})_y[\mathbf{n}\cdot(\mathbf{e}'\times\mathbf{e})]\}f(\delta,\mathbf{q}), \quad (10b)$$

$$f(\delta,\mathbf{q})=ku\sqrt{2/\pi}\int d\mathbf{p}\,W(\mathbf{p})\{\gamma_t-i[\delta-\omega(\mathbf{p}+\hbar(\mathbf{k}'-\mathbf{q}),\mathbf{p}+\hbar(\mathbf{k}-\mathbf{q}))]\}^{-1}, \quad (10c)$$

and l is the length over which the probe interacts with the atoms. Corrections to $(\mathbf{E}'_{\text{out}})_x$ have been neglected. In the expression

$$\omega[\mathbf{p}+\hbar(\mathbf{k}'-\mathbf{q}),\mathbf{p}+\hbar(\mathbf{k}-\mathbf{q})]=(\mathbf{k}'-\mathbf{k})\cdot\mathbf{p}/m-\hbar(\mathbf{k}'-\mathbf{k})\cdot\mathbf{q}/m, \tag{11}$$

the first term is responsible for Doppler broadening of the two-quantum transition, and the second term corresponds to the shift of the line caused by recoil during spontaneous emission. Assuming that the recoil shift is small, one finds that, for the geometry of Fig. 1,

$$\epsilon=(\pi^{3/2}/45\sqrt{2})(N|\mu|^2 l/\hbar u)[|\chi|^2\Gamma/(\gamma^2+\Delta^2)(\gamma-i\Delta)](\omega_k/ku)^2 w''[(\delta-i\gamma_t)/\sqrt{2}ku], \tag{12}$$

where $\omega_k=\hbar k^2/2m$ is a recoil frequency, $w(z)=(imu/\sqrt{\pi})\int dt\ W_1(mut)/(z-t)$, and it has been assumed that $W(\mathbf{p})=W_1(p_x)W_1(p_y)W_1(p_z)$.

For complex ϵ, the output field (10a) is elliptically polarized (see Fig. 2). The angle of Faraday rotation θ and the ratio b_2/b_1 of the ellipse half-axes are given by [8]

$$\theta=\mathrm{Re}(\epsilon), \quad b_2/b_1=\mathrm{Im}(\epsilon). \tag{13}$$

The RIOFR for $\Delta=0$ as a function of δ is shown in Fig. 3.

In estimating the magnitude of the effect, one notices that for the cases of exact resonance, $\Delta=0$, where the effect is maximum, there is no dependence on the dipole moment matrix element in Eq. (12). As a result one finds that, in the Doppler limit $\gamma_t \ll ku$, the amplitude $\Delta\theta=\theta_{\max}-\theta_{\min}$ in Fig. 3 is given by

$$\Delta\theta \approx 9.9\times10^{-4}[\omega_k^2/(ku)^3]S\lambda^5 Nl/\hbar c, \tag{14}$$

where $S=c|E|^2/8\pi$ is the pump field Poynting vector and $\lambda=2\pi/k$. For typical values $\omega_k\sim10^5\ \mathrm{s}^{-1}$, $\omega_k/ku\sim0.1$, $S\sim10^{-4}\ \mathrm{W/cm^2}$, $\lambda\sim1\mu$, $N\sim10^9\ \mathrm{cm}^{-3}$, and $l\sim0.1$ cm, one finds that $\Delta\theta\sim3\times10^{-4}$ rad.

III. DISCUSSION

The results obtained above can be given a simple geometric interpretation. Consider first the dynamical part of the problem. One can show that, for the chosen geometry, no OFR can occur in the absence of recoil. The spin of the excited or ground state varies as $\mathbf{s}_{ii}\propto\mathbf{e}'\times\mathbf{e}$, since $\mathbf{e}'\times\mathbf{e}$ is the only axial vector one can construct from polar vectors $\mathbf{e}'$ and $\mathbf{e}$. In contrast to the pseudovectors $\mathbf{s}_{ii}$, the "spin" of the coherence $\mathbf{s}_{21}$ is a polar vector, because it is a matrix element of the spin between states $|1\rangle$ and $|2\rangle$, having opposite parity. This vector determines the gas polarization $\mathbf{P}_+$[see Eq (8)]. When the pump field scatters from the ground- or excited-state gratings, the only possible polar vector one can construct is $\mathbf{P}_+\propto\mathbf{e}\times(\mathbf{s}_{22}-\mathbf{s}_{11})$. As a consequence, the stimulated part of the medium's polarization varies as $\mathbf{P}_+^{st}\propto\mathbf{e}\times(\mathbf{e}\times\mathbf{e}')=\mathbf{e}(\mathbf{e}\cdot\mathbf{e}')-\mathbf{e}'$. This vector is in the probe field's plane of polarization $[(\mathbf{P}_+^{st})_y=0]$; no OFR occurs.

The situation changes when spontaneous decay is included. The probability of spontaneous transition is bilinear in the photon polarization vector $\mathbf{e}^{\lambda}(\mathbf{q})$. The sum over polarizations can be carried out using the relation $\Sigma_{\lambda=1,2}e_i^{\lambda}(\mathbf{q})e_k^{\lambda}(\mathbf{q})=\delta_{ik}-q_iq_k/q^2$. One can conclude from this formula that, when the excited-state grating $\mathbf{s}_{22}$ decays, the ground-state grating $\mathbf{s}_{11}$ is also bilinear in $\mathbf{q}$. For a pseudovector $\mathbf{s}_{11}$ one can expect two bilinear combinations, $\mathbf{s}_{11}\propto\mathbf{q}(\mathbf{q}\cdot\mathbf{s}_{22})$ or $\mathbf{s}_{11}\propto\mathbf{s}_{22}\mathbf{q}^2$. The second combination is insensitive to the spontaneous photon's propagation direction and can lead only to the same contribution as stimulated

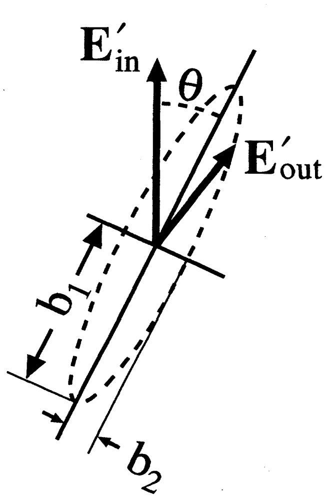

FIG. 2. An incident linearly polarized probe field $\mathbf{E}'_{\text{in}}$ is transformed into an elliptically polarized field with its major axis rotated by angle θ when it exits the sample.

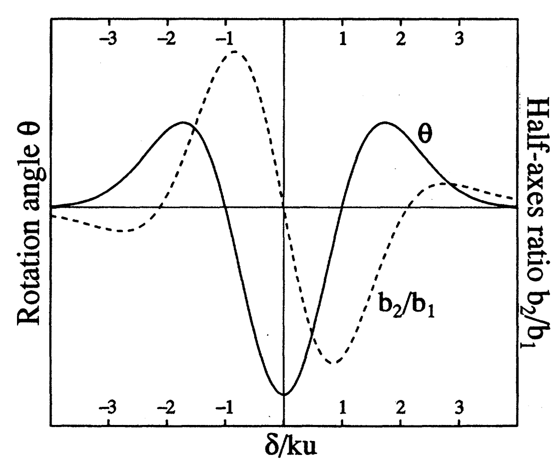

FIG. 3. Frequency dependence $[\delta/ku=(\Omega'-\Omega)/ku]$ of the angle of rotation (solid curve) and the ratio of the polarization ellipse half-axes (dashed curve) for a pump field tuned on resonance ($\Delta=0$). For the parameters chosen in the text, $\theta_{\max}-\theta_{\min}\simeq3.0\times10^{-4}$ and $(b_2/b_1)_{\max}\simeq1.7\times10^{-4}$. It is assumed that $\gamma_t/ku\ll1$, $W_1(p)=(1/\sqrt{\pi}\ mu)\exp[-(p/mu)^2]$.

processes. Thus, the term of interest is $\mathbf{s}_{11} \propto \mathbf{n}(\mathbf{n}\cdot\mathbf{s}_{22}) \propto \mathbf{n}[\mathbf{n}\cdot(\mathbf{e}'\times\mathbf{e})]$ (where $\mathbf{n}=\mathbf{q}/q$), which leads to a contribution to the polarization proportional to the vector quantity

$$\mathbf{A}=(\mathbf{e}\times\mathbf{n})[\mathbf{n}\cdot(\mathbf{e}'\times\mathbf{e})]. \tag{15}$$

The y component of this vector can be identified in Eq. (10b). For $\mathbf{e}'=(1,0,0)$, $\mathbf{e}=(e_x,0,e_z)$ one finds

$$A_y=(e_x n_z - e_z n_x) n_y e_z . \tag{16}$$

This expression still has a zero mean value $\langle A_y\rangle_{\mathbf{n}}=0$.

Before averaging over various directions $\mathbf{n}$, one must multiply A_y by an appropriate line-shape factor. To understand the manner in which the line-shape factor arises, we consider a process in which (i) pump and probe fields create a spatial grating of excited-state spin $\mathbf{s}_{22}$; (ii) this spin decays via spontaneous emission, inducing a grating of ground-state spin $\mathbf{s}_{11}$; (iii) the pump field scatters from $\mathbf{s}_{11}$, leading to the desired polarization (9).

In the first step, starting from a spatially uniform initial density matrix $|\mathbf{p}\rangle\langle\mathbf{p}|$ in the ground state, the fields produce a coherent mixture of excited states $|\mathbf{p}+\hbar\mathbf{k}'\rangle\langle\mathbf{p}+\hbar\mathbf{k}|$ having spin (4). This excited-state spin varies as $\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}-i\delta t]$.

In the second step, spontaneous decay, in turn, transfers the excited-state grating to a long-lived ground-state density-matrix grating, which is a coherent mixture of momenta $\mathbf{p}+\hbar(\mathbf{k}'-\mathbf{q})$ and $\mathbf{p}+\hbar(\mathbf{k}-\mathbf{q})$, modified by the momentum $\hbar\mathbf{q}$ of the emitted photon. This overall process is resonant for a pump-probe detuning

$$\delta=(\mathbf{k}'-\mathbf{k})\cdot\mathbf{p}/m-\hbar(\mathbf{k}'-\mathbf{k})\cdot\mathbf{q}/m, \tag{17}$$

which determines the center of the narrow two-quantum line [see Eqs. (10c) and (11)]. The line-shape factor depends on the recoil shift

$$\omega_{\mathbf{q}}=2\omega_k(\mathbf{n}_{\mathbf{k}'}-\mathbf{n}_{\mathbf{k}})\cdot\mathbf{n}, \tag{18}$$

where $\mathbf{n}_{\mathbf{k}'}$ and $\mathbf{n}_{\mathbf{k}}$ are unit vectors along $\mathbf{k}'$ and $\mathbf{k}$, respectively. Since $(\mathbf{k}'-\mathbf{k})$ has a nonzero projection on the y axis, contributions to the medium's response accompanied by photon emission to the right and left sides of the probe field's plane of polarization are not equal. This asymmetry leads to a macroscopic optical activity of the vapor and to the RIOFR. When the line-shape factor is expanded to order $\omega_{\mathbf{q}}^2$, one finds a contribution to the atomic polarization $(\mathbf{P}_+)_y$, which varies as

$$\langle A_y n_z n_y\rangle = e_x e_z \langle n_y^2 n_z^2\rangle_{\mathbf{n}} \neq 0, \tag{19}$$

which leads to the optical Faraday rotation.

As was mentioned above, one can exclude stimulated contributions to the OFR when the pump field polarization vector $\mathbf{e}$ is in the (y,z) plane, since $\mathbf{e}'\cdot\mathbf{e}=0$. In this case RIOFR also occurs. If, for example, the pump field propagates along the x axis, a similar analysis shows that the medium's polarization is proportional to $e_z e_y \langle n_x^2 n_y^2\rangle_{\mathbf{n}}$.

Although the discussion was given for a $J=\frac{1}{2}$ to $J'=\frac{1}{2}$ transition, the qualitative nature of the results is unchanged for different values of J and J' (provided $J\neq 0$ and $J'\neq 0$). It is the polarization and propagation directions of the fields that determine whether or not the medium is optically active. For our chosen geometry, the stimulated contribution to OFR varies as $e_x e_y$ and the contribution to $(\mathbf{P}_+)_y$ is given by Eq. (19), independent of J and J'.

There is, however, an important difference between the RIR and RIOFR. The observation of the RIR is invariably related to a measure of the total ground-state population [9], which remains constant in the absence of recoil. The decay time γ_t associated with the total population is typically much smaller than the optical pumping rates, allowing for very narrow RIR and ratios of RIR signal to background that are much greater than unity. On the other hand, the γ_t appearing in this paper is associated with the decay of ground-state Zeeman coherence, which is typically determined by the optical pumping rates. As such it is possible to have situations in which $\gamma_t \geqslant ku$; in this limit the line shape for RIOFR is proportional to the second derivative of a Lorentzian having width $2\gamma_t$. It should still be possible to measure a RIOFR since the signal appears on a "black" background for properly chosen field geometries. To properly account for optical pumping and any effects of spatial localization, a full quantum calculation is needed.

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