be possible to produce a substantial population in a single vibrational-rotational level of a Rydberg state. Phenomena such as superradiant or microwave transitions, autoionization, and field ionization could then be studied.

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Linear Electro-Optic Effect in Gases

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The *linear* effect of an electrostatic field on the intensity of scattering of light by gas is reported. The change in the depolarized intensity scattered by gaseous methyl chloride in the y direction from a beam of right circularly polarized light propagating in the z direction due to an electrostatic field $F_x = 7\times 10^6$ V m^{-1} is -7 ± 1 ppm. It changes sign on reversing F_x, the direction of scattering, and the circularity of the light. The effect should be exhibited by all materials.

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Crystals may exhibit linear electro-optical properties, but gases and liquids are expected to show quadratic effects of an electric field on their optical properties. However, we report a linear effect of an electric field on the intensity of light scattered from a circularly polarized beam by gaseous methyl chloride; the differential scattering is of opposite sign in right- and left-circular polarization (Fig. 1).

In 1846 Faraday observed a linear effect of a magnetic field on optical rotation.[1] Magneto-optical rotation was later shown to be a property possessed by all materials. We believe that all substances also have a linear electro-optical scattering power.[2]

Figure 2 shows the apparatus. Argon-laser radiation at 488 nm was circularly polarized by a Pockels cell and focused into the cell. An elec-

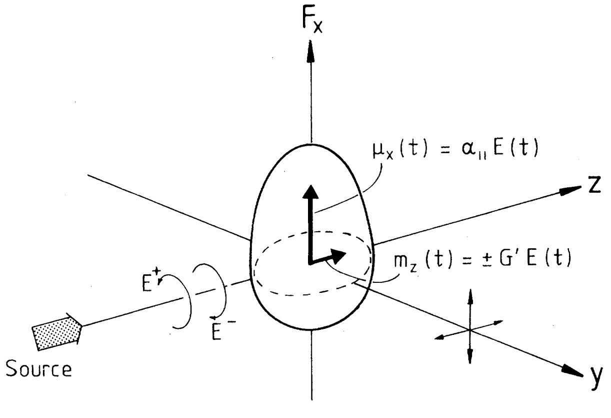

FIG. 1. A polar molecule such as CH_3Cl oriented by the electric field F_x. The scattering from right (E^+) and left (E^-) circularly polarized light with x or z polarization is observed on the y axis.

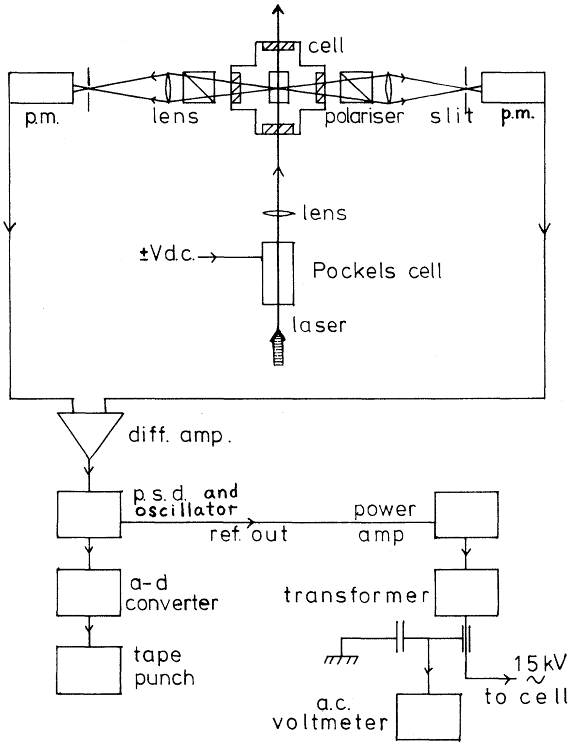

FIG. 2. A block diagram of the apparatus.

tric field $F_x = F_x^{(0)}\cos(2\pi f t)$, where $f = 170$ Hz was applied to stainless-steel electrodes 2.3 mm apart. Light scattered in the $\pm y$ directions was collected by two photomultipliers. Their outputs were *subtracted* and analyzed with a phase-sensitive detector. The signal was fed to an analog-to-digital converter and put onto paper tape. After about a minute, the circular polarization was changed by reversing the voltage on the Pockels cell and the process repeated. The changes ΔI_z^+ and ΔI_z^- in the depolarized intensities scattered from right and left circularly polarized beams were found to vary linearly with F_x and with the pressure of CH_3Cl. Fifty pairs of measurements were made on CH_3Cl at room temperature and 3.5 atm with $F_x^{(0)} = 7\times10^6$ V m^{-1}, giving

$$\frac{\Delta I_z^+}{I_z^+} = -\frac{\Delta I_z^-}{I_z^-} = (-1.0\pm0.1)F_x/(10^{12}\ \mathrm{V\ m^{-1}}). \tag{1}$$

To interpret the phenomenon, it is necessary to consider the oscillating electric and magnetic dipoles induced in a molecule by the optical field.[2] Natural optical activity results from the trace of the tensor $G_{\alpha\beta}'$ giving the electric dipole induced by $\dot{\vec{B}}(t)$, the time derivative of the magnetic vector of the electromagnetic field, and the magnetic dipole $\vec{m}(t)$ induced by $\dot{\vec{E}}(t)$, the time derivative of the electric field[3]:

$$\mu_\alpha(t) = \alpha_{\alpha\beta}E_\beta(t) + \alpha_{\alpha\beta}'\dot{E}_\beta(t)\omega^{-1} + G_{\alpha\beta}'\dot{B}_\beta(t)\omega^{-1}, \tag{2}$$

$$m_\alpha(t) = -G_{\beta\alpha}'\dot{E}_\beta(t)\omega^{-1}. \tag{3}$$

Here ω is the angular frequency of the optical field.

Since $\vec{\mu}$, $\vec{E}$, and $\dot{\vec{B}}$ are even under time reversal θ, while $\dot{\vec{E}}$, $\vec{m}$, and $\vec{B}$ are odd, the symmetric polarizability tensor $\alpha_{\alpha\beta}$ and $G_{\alpha\beta}'$ are even under θ. Under parity P, $\vec{\mu}$ and $\vec{E}$ are odd, but $\vec{m}$ and $\vec{B}$ are even, so that $\alpha_{\alpha\beta}$ is even and $G_{\alpha\beta}'$ is odd. The Faraday rotation comes from the linear effect of a magnetostatic field[4] B_z on $\alpha_{\alpha\beta}'$:

$$\alpha_{\alpha\beta}'(B) = \alpha_{\alpha\beta}' + \alpha_{\alpha\beta\gamma}'^{(B)}B_\gamma. \tag{4}$$

The third-rank tensor $\alpha_{\alpha\beta\gamma}'^{(B)}$ has an isotropic part which gives different refraction for right- and left-circular polarization in fluids. It is even under θ and P and therefore exists for all molecules.

The new phenomenon comes from the linear effect of F_x on the optical-activity tensor[5] $G_{\alpha\beta}'$:

$$G_{\alpha\beta}'(F) = G_{\alpha\beta}' + G_{\alpha\beta\gamma}'^{(F)}F_\gamma. \tag{5}$$

Like $\alpha_{\alpha\beta\gamma}'^{(B)}$, $G_{\alpha\beta\gamma}'^{(F)}$ is even under θ and P and therefore exists for all materials.

The effect may be understood with aid of Fig. 1. We consider an orientated, axially symmetric molecule whose polarizability has two independent components $\alpha_{xx} = \alpha_\parallel$ and $\alpha_{yy} = \alpha_{zz} = \alpha_\perp$; $G_{\alpha\beta}'$ has just one, $G_{yz}' = -G_{zy}' = G'$, where x is the molecular axis of symmetry[2,3]:

$$\alpha_{\alpha\beta} = \sum_{j\neq n}\frac{2\omega_{jn}\,\mathrm{Re}\{\langle n|\mu_\alpha|j\rangle\langle j|\mu_\beta|n\rangle\}}{\hbar(\omega_{jn}^2-\omega^2)} \tag{6}$$

$$G_{\alpha\beta}' = -\sum_{j\neq n}\frac{2\omega\,\mathrm{Im}\{\langle n|\mu_\alpha|j\rangle\langle j|m_\beta|n\rangle\}}{\hbar(\omega_{jn}^2-\omega^2)}. \tag{7}$$

Polarized scattering, with electric vector in the x direction, comes from the oscillating dipoles $\mu_x(t)$, $m_z(t)$. For right ($\vec{E}^+$) and left ($\vec{E}^-$) circularly polarized incident radiation propagating in the z direction,

$$\begin{aligned} E_x^\pm(t) &= E^{(0)}\cos\omega(t-zc^{-1}), \\ E_y^\pm(t) &= \mp E^{(0)}\sin\omega(t-zc^{-1}). \end{aligned} \tag{8}$$

There is an oscillating magnetic dipole [see Eq.

(3) and Fig. 1]

$$m_z(t) = -G'\dot{E}_y(t)\omega^{-1} = \pm G'E^{(0)}\cos\omega(t - zc^{-1}), \tag{9}$$

and its radiation interferes with that from

$$\mu_x(t) = \alpha_\parallel E_x(t) = \alpha_\parallel E^{(0)}\cos\omega(t - zc^{-1}) \tag{10}$$

leading to opposite differential scattering in right and left circularly polarized light.

To understand differential *depolarized* scattering it is necessary to consider an incompletely orientated molecule. The scattering is due to $\mu_z(t)$ and $m_x(t)$:

$$\mu_z(t) = \alpha_{zz}E_x(t) + \alpha_{zy}E_y(t), \tag{11}$$

$$m_x(t) = -G_{yx}'\dot{E}_y(t)\omega^{-1} = \pm G_{yx}'E^{(0)}\cos\omega(t - zc^{-1}). \tag{12}$$

For a molecule such as CH_3Cl (with unit vector $\vec{n}$ along the dipole axis)

$$\mu_z(t) = (\alpha_\parallel - \alpha_\perp)E^{(0)}[n_z n_x\cos\omega(t - zc^{-1}) \mp n_z n_y\sin\omega(t - zc^{-1})], \tag{13}$$

$$m_x(t) = \mp G' n_z E^{(0)}\cos\omega(t - zc^{-1}). \tag{14}$$

There is differential scattering $\Delta I_z^\pm$ resulting from interference in the radiation from the magnetic dipole and the first part of $\mu_z(t)$. In the field F_x, $\langle n_x n_z^2\rangle = \mu_0 F_x/15kT$, where μ_0 is the permanent dipole of the permanent dipole of the molecule, and

$$\frac{\Delta I_z^+}{I_z^+} = -\frac{\Delta I_z^-}{I_z^-} = \frac{2c^{-1}\langle\mu_z(t)m_x(t)\rangle}{\langle\mu_z^2(t)\rangle} = \frac{G'\mu_0 F_x}{c(\alpha_\parallel - \alpha_\perp)kT}. \tag{15}$$

The expected magnitude of the differential scattering induced by F_x is of the order of a few parts in a million, and polar molecules should show larger effects than nonpolar ones.[2]

For CH_3Cl, $\mu_0 = 1.87$ D $= 6.24\times10^{-30}$ C m and $\alpha_\parallel - \alpha_\perp = 1.768\times10^{-40}$ C^2 m^2 J^{-1},[6] and so from Eqs. (1) and (15), $G' = (-34\pm4)\times10^{-36}$ C^2 m^3 s^{-1} J^{-1} (the positive axis is from Cl to CH_3).

Optical rotation is determined by the symmetric part of $G_{\alpha\beta}'$ and differential scattering by the antisymmetric part. $G_{\alpha\beta}'$ reflects the *distribution* of polarizable matter in the molecule. The value of G' for CH_3Cl shows that when an oscillating electric field is applied at right angles to the C-Cl axis the oscillating dipole is located more on the Cl than the CH_3. If the transition dipoles in (7) were 1 D and 1 μ_B and the excitation energy $\hbar\omega_{jn}$ $\times10^{-18}$ J (50 000 cm^{-1}), G' would be -30×10^{-36} C^2 m^3 s^{-1} J^{-1}.

If the effect were studied near absorption frequencies one could expect to obtain information about optical properties of atoms and molecules in excited electronic states.

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