

## Magnetolectric Jones Dichroism in Atoms

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The authors suggest that atomic experiments measuring the interference between magnetic-dipole and electric-field-induced electric-dipole transition amplitudes provide a valuable system to study magnetolectric Jones effects.

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A recent Letter [1] reported the first observation of magnetolectric Jones birefringence in liquids (see also Ref. [2]). This observation helped to clarify some of the long-standing theoretical confusion surrounding Jones birefringence and the associated Jones dichroism (collectively known as Jones effects) [3,4]. The interest in further understanding these effects has led to the investigation of other experimental systems which may exhibit Jones effects. These include the possibility of observing the effects through beam divergence in uniaxial crystals [5] and possible observation in the quantum vacuum [6]. In this Letter we point out that Jones dichroism can be studied in atomic systems under much less severe experimental requirements. In addition, these atomic systems are more amenable to theoretical analysis than the relatively complicated condensed-matter systems that have been studied to date. The simplicity of these systems may help to expand the understanding of the manifestation of Jones effects in general. We also point out that our recent experiment [7] measuring interference between magnetic-dipole and electric-field-induced electric-dipole transition amplitudes in atomic ytterbium constitutes a measurement of Jones dichroism in a simple atomic system.

The development of the Jones matrix calculus for describing the propagation of light led to the prediction of two distinct types of linear birefringence and dichroism [8]. The two types of effects differ in the orientation of the birefringent and dichroic axes. The Jones formalism revealed that certain uniaxial media may exhibit birefringence and dichroism along axes which are at  $\pm 45^\circ$  relative to the axis of anisotropy. Birefringent and dichroic effects of this type are called Jones effects. They are distinct from the familiar birefringence and dichroism, which have axes parallel and perpendicular to the axis of anisotropy.

There has been theoretical discussion concerning the requirements for media that may exhibit Jones effects and what transition moments must be accounted for in order to describe the effects [3,4]. In Ref. [4], it was shown that Jones effects may be induced in isotropic media by the application of parallel electric and magnetic fields. If the direction of light propagation is perpendicular to

the electric and magnetic fields, the Jones effects are described by

$$\Delta n_J \equiv n_{+45^\circ} - n_{-45^\circ}, \quad (1)$$

where  $n_{\pm 45^\circ}$  is the complex index of refraction for light polarized at  $\pm 45^\circ$  relative to the electric and magnetic fields. The real and imaginary parts of  $\Delta n_J$  describe Jones birefringence and dichroism, respectively. On a microscopic level, Jones dichroism may manifest itself as a difference between the rates with which atoms of the medium are transferred to an excited state in the presence of light polarized at  $\pm 45^\circ$  relative to the electric and magnetic fields given by

$$\Delta \Gamma_J \equiv \Gamma_{+45^\circ} - \Gamma_{-45^\circ}. \quad (2)$$

Jones effects generally occur in materials which exhibit the more familiar birefringence and dichroism. In addition, Jones effects are predicted to be significantly smaller than the usual birefringence and dichroism in most media. Consequently, magnetolectric Jones birefringence has been observed only recently in molecular liquids under extreme experimental conditions [1,2].

To our knowledge, the observation of Jones dichroism has not been reported as such. Here we point out that Stark-interference experiments [9] which utilize parallel electric and magnetic fields provide a simple atomic system which exhibits Jones dichroism. The experiment [7] measuring a highly forbidden magnetic-dipole transition amplitude in atomic ytterbium using this technique constitutes such a system and its results can be interpreted as an observation of magnetolectric Jones dichroism.

In the experiment [7], we studied a highly forbidden transition between states of the same parity. In the absence of external fields and neglecting parity-nonconserving effects, the transition occurs only through a small magnetic-dipole amplitude ( $\approx 10^{-4} \mu_B$ , where  $\mu_B$  is the Bohr magneton). By applying a static electric field, an electric-dipole amplitude is induced through mixing of opposite-parity states. An atomic beam of ytterbium was excited with resonant laser light propagating perpendicularly to parallel electric and magnetic fields. The excitation light was polarized at an angle  $\theta$

relative to the external fields. For the transition studied in our experiment (between a ground state with total angular momentum equal to zero and an excited state with total angular momentum equal to one), the electric field  $\mathbf{E}$  results in a Stark-induced electric-dipole transition amplitude to the  $M'_J$  magnetic sublevel of the excited state given by [9]

$$A(E1_{\text{St}}) = i\beta(\mathbf{E} \times \boldsymbol{\varepsilon})_{-M'_J}, \quad (3)$$

where  $\boldsymbol{\varepsilon}$  is the electric-field amplitude of the laser light,  $(\mathbf{E} \times \boldsymbol{\varepsilon})_{-M'_J}$  is the  $-M'_J$  component of the vector in the spherical basis, and  $\beta$  is the vector transition polarizability [10]. The magnetic-dipole transition amplitude is given by

$$A(M1) = \mu(\hat{\mathbf{k}} \times \boldsymbol{\varepsilon})_{-M'_J}, \quad (4)$$

where  $\hat{\mathbf{k}}$  is the direction of propagation of the excitation light,  $\hat{\mathbf{k}} \times \boldsymbol{\varepsilon}$  is the magnetic-field amplitude of the light, and  $\mu$  is the magnetic-dipole matrix element between the ground state and any of the  $M'_J$  magnetic sublevels of the excited state. The transition rate is therefore

$$\begin{aligned} \Gamma &\propto \sum_{M'_J} |A(E1_{\text{St}}) + A(M1)|^2 \\ &\propto \sum_{M'_J} |A(E1_{\text{St}})|^2 + 2\text{Re}[A(E1_{\text{St}})A(M1)^*] + |A(M1)|^2. \end{aligned} \quad (5)$$

As discussed in Ref. [7], the interference term in Eq. (5) is of opposite sign for the  $M'_J = +1$  and  $M'_J = -1$  magnetic sublevels. It is therefore necessary to apply a magnetic field to resolve the different sublevels in order to observe the effect of this term. The signal due to the interference term is proportional to the rotational invariant

$$[(\mathbf{E} \times \boldsymbol{\varepsilon}) \times (\hat{\mathbf{k}} \times \boldsymbol{\varepsilon})] \cdot \hat{\mathbf{B}}. \quad (6)$$

This is also true in a more general case where  $\mathbf{E}$  and  $\mathbf{B}$  are not necessarily collinear.

We define the  $z$  axis to be along the direction of the magnetic field,  $\mathbf{B} = B\hat{\mathbf{z}}$ , and define the  $x$  axis so that the electric field lies in the  $x$ - $z$  plane,  $\mathbf{E} = E_x\hat{\mathbf{x}} + E_z\hat{\mathbf{z}}$ . We assume that the light propagation is perpendicular to both fields,  $\mathbf{k} = k\hat{\mathbf{y}}$ , and that the light is linearly polarized at an angle  $\theta$  relative to the magnetic field,  $\boldsymbol{\varepsilon} = \sin\theta\hat{\mathbf{x}} + \cos\theta\hat{\mathbf{z}}$  (Fig. 1). The  $|A(E1_{\text{St}})|^2$  and  $|A(M1)|^2$  terms in Eq. (5) are independent of the sign of the angle of polarization while the interference term is odd with  $\theta$ . Using expression (6) it is easily shown that the difference in transition rates for  $\pm\theta$  results in a Jones dichroism given by

$$\Gamma_{+\theta} - \Gamma_{-\theta} \propto E_z \sin\theta \cos\theta. \quad (7)$$

The factor  $E_z$  in Eq. (7) shows that

$$\Delta\Gamma_J \propto \mathbf{E} \cdot \hat{\mathbf{B}}, \quad (8)$$

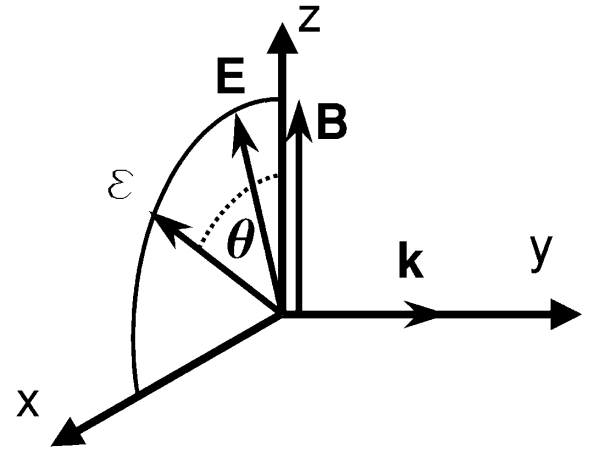


FIG. 1. Orientation of external fields.

which is the predicted dependence of the magnetoelectric Jones effects on  $\mathbf{E}$  and  $\mathbf{B}$  [4]. It is interesting to note that the transition rate depends on the magnitude of the magnetic field only for values of the magnetic field which do not fully resolve the magnetic sublevels. This is analogous to the change in magnetic-field dependence of the resonant Faraday rotation (see, for example, Ref. [11]).

Because of the weakness of the forbidden transition studied, we determined the transition rate by observing fluorescence in a decay branch of the excited state rather than detecting absorption. In our experiment the observed Jones dichroism is significantly smaller ( $\approx 5 \times 10^{-3}$  at the electric fields used in the experiment) than the normal dichroism, which is dominated by the Stark-induced amplitude. As can be seen from Eq. (3), only the component of the light electric field that is perpendicular to  $\mathbf{E}$  contributes to the transition rate. Thus, the dominant fluorescence signal is proportional to  $\sin^2\theta$  and to  $|E|^2$ . These dependences were verified experimentally. The interference term responsible for Jones dichroism was isolated from the dominant signal by comparing the fluorescence spectra for opposite electric fields. In the data analysis we normalized the interference term to the dominant signal resulting in an asymmetry given by

$$\frac{\Gamma(E_+) - \Gamma(E_-)}{\Gamma(E_+) + \Gamma(E_-)} = \frac{2M1 \cos(\theta)}{\beta E \sin(\theta)} M_J. \quad (9)$$

The dependence of the asymmetry on the electric field, magnetic field, and polarization angle was verified experimentally (see Ref. [7] for a figure showing the interference term versus the magnitude of the electric field). Figure 2 shows the experimental fractional asymmetry [Eq. (9)] plotted versus the polarization angle. We have normalized the signal to the magnitude of the electric field in order to combine data taken at different electric fields and leave only the polarization angle dependence. Also shown is the expected angular dependence of the asymmetry. Most of the data were taken with light

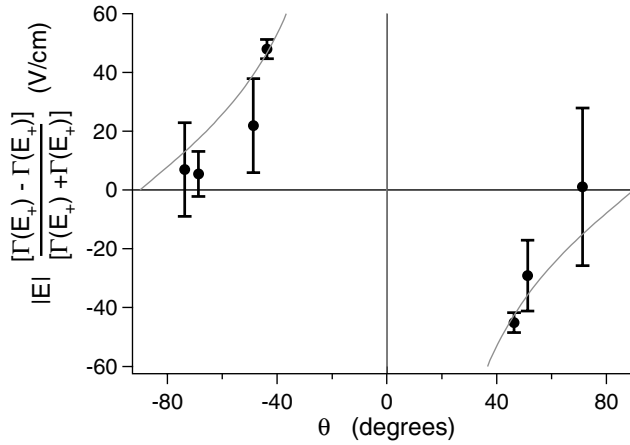


FIG. 2. Experimental results showing the dependence of the fractional transition-rate asymmetry, normalized by the magnitude of the electric field, on the angle of light polarization. The solid line shows the expected dependence. Data were taken in the work of Ref. [7] and experimental details are contained therein.

polarized at  $\theta = \pm 45^\circ$  relative to the electric field since the interference term is maximal at these values [see Eq. (7)]. The difference in the sign of the asymmetry for  $\theta = \pm 45^\circ$  clearly indicates a nonzero value of  $\Delta\Gamma_J$ , verifying the key signature of Jones dichroism.

We note that, although the Jones dichroism was significantly smaller than the usual dichroism in our experiment, it is possible to significantly increase its size by using an allowed magnetic-dipole transition. In fact, it is possible to have both the Jones dichroism and the regular dichroism of the same order as the overall absorption, which can be substantial in the case of an allowed magnetic-dipole transition.

Finally, we point out that atomic systems may be of use in measuring other types of magnetoelectric effects which are currently being studied in more complicated systems, such as more common forms of magnetoelectric linear birefringence [12] and magnetoelectric directional anisotropy [13]. In fact, expression (6) shows that this system exhibits both of these effects. It is interesting to note that a polarization-dependent directional anisotropy is present for both parallel and perpendicular electric and

magnetic fields. For the case of perpendicular electric and magnetic fields a component of the directional anisotropy is present even when averaged over the polarization angle.

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