COMMENTS

Coherent Production of Different m_L States in a Beam-Foil Experiment*

T. G. Eck
Department of Physics, Case Western Reserve University, Cleveland, Ohio 44106
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A simple model is presented in which the coherence between different m_L states in a tilted-foil, beam-foil experiment is induced by the electrostatic interaction between each beam particle and the foil as a whole. Agreement between the predictions of this model and the experimental results of Berry *et al.* seems sufficient to warrant refinement of the model and further experiments.

In beam-foil spectroscopic experiments, atoms or ions in excited states are produced by passing a high-speed ion beam through a thin foil, usually of carbon. If the plane of the foil is perpendicular to the direction of the incident ion beam, the cylindrical symmetry of the experiment about this direction implies the absence of any coherence in the radiations from excited states with different magnetic quantum numbers m_J ($=m_L+m_S$) along the beam.[1] If, further, Russell-Saunders (L-S) coupling is valid for these states, coherence can occur only between states with the same m_L and m_S. However, this symmetry can be destroyed by tilting the plane of the foil. In a recent beam-foil experiment Berry, Curtis, Ellis, and Schectman[2] have investigated coherence between the different m_L states of the $3p\,^1P$ term of ^{4}He by measuring the anisotropy of the 5016 Å radiation ($3p\,^1P$ to $2s\,^1S$ transition) as a function of the angle between the normal to the foil and the direction of the incident He^+ beam. The purposes of the present Comment are to present what I consider to be a plausible model for the physical origin of the coherence between different m_L states in such an experiment and to compare the radiation anisotropy predicted by this model with the data of Ref. 2. This work is part of an investigation of the whole question of coherent production of atomic states in beam-foil experiments, the details of which will be published elsewhere.[3]

The essential elements of the model are the assumptions that the beam particles exit from the final surface of the foil in states which, when averaged over many beam particles, are incoherent superpositions of the various m_L states *no matter how the foil is oriented,* and that the coherence between different m_L states is induced by the *electrostatic* interaction between each particle and the foil as a whole experienced by that particle while it is in the immediate vicinity of the foil. These assumptions seem plausible and have the added attraction of allowing one to proceed with the calculation of the dependence of coherence on foil orientation and particle speed.

A neutral atom will see an image dipole electric field which is very strong (of the order of 10^8 V/cm) when the atom is within a few Bohr radii of the foil, but falls off rapidly with distance from the foil. This strong electric field, *directed along the normal to the foil,* will remove the energy degeneracy between states of different m_L by Stark coupling to distant electronic states. If for the Stark-coupling calculation we choose our axis of quantization along the foil normal, the degeneracy between states whose m_L values differ only in sign will not be removed. These energy differences lead to the development of phase differences between the various m_L states as the particle traverses the region of large electric field. It is these phase differences, the same for each beam particle, which determine the coherence of different m_L states in the region beyond the foil.

For a $^1P_1$ state with no hyperfine structure the initial wave function just as the atom exits from the foil is

$$\psi_0 = f_1|1\rangle + f_0|0\rangle + f_{-1}|-1\rangle,$$

where f_i is the probability amplitude for produc-

ing the ith m_L state and the axis of quantization is along the beam. The amplitudes are related to the production cross sections σ_i by $\sigma_i = \langle |f_i|^2 \rangle$, where the angular brackets denote an average over beam particles. The lack of coherence between the initial m_L states means that $\langle f_i f_j^* \rangle = 0$ for $i \neq j$. To simplify the calculation of the effect of the surface interaction, we transform ψ_0 to an axis of quantization along the normal to the foil. Figure 1 of Ref. 2 shows that the foil coordinate system (foil normal along z') is obtained from the laboratory system (beam along z) by rotating counterclockwise about the x axis through the tilt angle α so that $x = x'$, $y = y' \cos\alpha - z' \sin\alpha$, and $z = y' \sin\alpha + z' \cos\alpha$. It is an elementary exercise in the transformation of angular momentum eigenfunctions to express ψ_0 in terms of the z' eigenfunctions to give

$$\psi_0' = a_1' |1\rangle' + a_0' |0\rangle' + a_{-1}' |-1\rangle',$$

with the a_i''s functions of the f_i's and the angle α.

The electrostatic interaction lifts the degeneracy between $|0\rangle'$ and the states $|1\rangle'$ and $|-1\rangle'$, but leaves $|1\rangle'$ and $|-1\rangle'$ degenerate. When the beam particle has passed beyond the influence of the foil, we have

$$\psi' = a_1' |1\rangle' + a_0' |0\rangle' \exp(-i\varphi) + a_{-1}' |-1\rangle',$$

where

$$\varphi = \int_{t_0}^{\infty} \omega(t)\, dt = (1/v \cos\alpha) \int_{z_0'}^{\infty} \omega(z')\, dz',$$

v is the particle speed, and ω is the time-varying angular frequency difference between $|0\rangle'$ and the degenerate states $|1\rangle'$ and $|-1\rangle'$. Taking the upper limit of the phase integral equal to ∞ causes negligible error since the magnitude of ω decreases rapidly with distance from the foil. The choice of the lower limit for this integral is much more problematic, reflecting the gross simplicity of the model adopted for the particle-foil interaction. We shall return to this point presently. Finally, we transform back to an axis of quantization along the beam to obtain a wave function which can be used to predict the radiation anisotropy for comparison with the results of Ref. 2. This gives

$$\psi = a_1 |1\rangle + a_0 |0\rangle + a_{-1} |-1\rangle, \tag{1}$$

with

$$\begin{aligned} a_0 &= -(i/\sqrt{2})(f_1 + f_{-1}) \sin\alpha \cos\alpha [1 - \exp(-i\varphi)] + f_0 [\sin^2\alpha + \cos^2\alpha \exp(-i\varphi)], \\ a_1 + a_{-1} &= (f_1 + f_{-1})[\cos^2\alpha + \sin^2\alpha \exp(-i\varphi)] + i\sqrt{2} f_0 \sin\alpha \cos\alpha [1 - \exp(-i\varphi)], \\ a_1 - a_{-1} &= f_1 - f_{-1}. \end{aligned} \tag{2}$$

Berry *et al.*[2] have characterized the anisotropy of the $3p\,{}^1P_1$ to $2s\,{}^1S_0$ radiation by determining the Stokes parameters[4] I, M, C, and S for radiation in the x-z plane at angles $\theta = 53°$ and $90°$ with the z axis. These parameters can be expressed in terms of $E_\parallel$ and $E_\perp$, the radiation electric field components parallel and perpendicular to the x-z plane (see footnote 12 of Ref. 2). In turn, $E_\parallel$ and $E_\perp$ can be expressed in terms of the a_i's and the angle θ. Since the expressions for the Stokes parameters are somewhat complicated for arbitrary θ, and the degree of agreement with the experimental results is the same for $\theta = 53°$ and $90°$, we limit our discussion to the case of $\theta = 90°$ (observation along the x axis). For this case

$$E_\parallel = -a_0, \quad E_\perp = -(i/\sqrt{2})(a_1 + a_{-1}), \tag{3}$$

where we have omitted constants of proportionality irrelevant to the present discussion. Equations (2) and (3) and footnote 12 of Ref. 2 yield

$$\begin{aligned} M/I &= R[1 - 2\sin^2(2\alpha) \sin^2(\varphi/2)], \\ C/I &= R \sin(4\alpha) \sin^2(\varphi/2), \\ S/I &= R \sin(2\alpha) \sin\varphi, \end{aligned} \tag{4}$$

where $R = (\sigma_0 - \sigma_1)/(\sigma_0 + \sigma_1)$ and use has been made of the fact that $\langle |f_{-1}|^2 \rangle = \langle |f_1|^2 \rangle \equiv \sigma_1$. The phase angle $\varphi = \varphi_0/\cos\alpha$, where φ_0 is the phase for $\alpha = 0$. Results identical to those of Eq. (4) are obtained if we start with Berry *et al.*'s Eq. (2) and use our Eqs. (1) and (2) to calculate the required expectation values of angular momentum operators. Equation (4) predicts a polarization fraction $f_p = (M^2 + C^2 + S^2)^{1/2}/I$ which is equal to $(\sigma_0 - \sigma_1)/(\sigma_0 + \sigma_1)$ for all values of α and φ. Thus, the surface interaction alters the nature of the polarization as the foil is tilted, but does not change its degree.

Berry *et al.*'s results have rather low precision, a reflection of the difficulty of these types of measurements. Within the stated error limits, the $\theta = 90°$ results in Table I of Ref. 2 are in good agreement with the present predictions for the dependence on α of M/I and S/I in the sense that these results are consistent with the single value for R and for φ_0. Also, the experimental polarization fraction is, within the error limits, independent of α. However, their results for C/I

are in rather violent disagreement with this paper's predictions. Equation (4) predicts that C/I should be positive for α between 0 and 45° and should vanish at $\alpha = 45°$. The experimental data yield a negative C/I with its maximum absolute value at 45°.

While for simplicity the present discussion so far has been carried out in terms of a very naive model which describes the particle-foil surface interaction via an electric field acting on an otherwise isolated atom, all that really seems to be required to obtain Eq. (4) is the assumption of an electrostatic, vector interaction whose direction is along the normal to the foil. All of the detailed physics of this interaction is contained in the phase φ_0. Questions such as the one raised above regarding the choice of a lower limit for the phase integral just point to the need for a more detailed picture of the interaction.

The agreement between the predictions of the present model and the experimental data, while by no means perfect, seems sufficient to warrant further investigation, both theoretical and experimental. The expressions for the Stokes parameters take on a particularly simple form for observation along the x axis if the two mutually perpendicular directions used in determining these parameters are taken along the foil and the normal to the foil (see Ref. 3). Since *any* mechanism where coherence is determined by the orientation of the foil is likely to yield its simplest results for this mode of observation, I recommend its use in future measurements of the kind carried out by Berry *et al.*

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[2]H. G. Berry, L. J. Curtis, D. G. Ellis, and R. M. Schectman, Phys. Rev. Lett. 32, 751 (1974).

[3]T. G. Eck, to be published.

[4]J. M. Stone, *Radiation and Optics* (McGraw-Hill, New York, 1963). See pages 309–320 for the relevant discussion.

Rotational-Vibrational Coupling in the Theory of Electron-Molecule Scattering

A. Temkin and E. C. Sullivan
Theoretical Studies Group, Goddard Space Flight Center, National Aeronautics Space Administration, Greenbelt, Maryland 20771
(Received 10 June 1974)

The adiabatic-nuclei approximation of vibrational-rotational excitation of homonuclear diatomic molecules can be simply augmented to describe the vibrational-rotational coupling by including the dependence of the vibrational wave function on j. Appropriate formulas are given and the theory is applied to e-H_2 excitation whereby it is shown that deviations from the simple Born-Oppenheimer approximation measured by Wong and Schultz can be explained. More important, it can be seen that the inclusion of the j-dependent centrifugal term is essential for transitions involving high-rotational quantum numbers.

The adiabatic-nuclei approximation[1] of rotational and/or vibrational excitation stems from the (approximate) separability of the total wave function Ψ of the electron-molecule system among rotational, vibrational, and electronic motion. In its simplest form this means

$$\Psi = \psi_{el}(\vec{r})\,\chi_v(R)\mathfrak{D}^{(j)}(\vec{\beta}). \tag{1}$$

In Eq. (1) $\vec{r}$ signifies the electronic coordinates, specifically, in the present context, those of the scattered electron; $\vec{R} = (R, \vec{\beta})$ are the internuclear coordinates with the angles $\vec{\beta}$ measured with respect to the incoming beam direction. The fundamental dynamical problem consists of determining ψ_{el}, and is done as a scattering problem in the fixed-nuclei approximation[2] giving rise to a fixed-nuclei amplitude[3]

$$f_R(\vec{\beta}, \Omega') = \sum a_{ll'm}(R)\mathfrak{D}_{m'm}^{(l)}(\vec{\beta})\mathfrak{D}_{0m}^{(l')*}(\vec{\beta})y_{lm'}(\Omega'), \tag{2}$$