Comment on elimination of polarization dependence from optical excitation functions

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The measurement of optical excitation functions excited by electron impact is typically accomplished by recording atomic fluorescence emitted into a small solid angle perpendicular to the incident electron beam. This measured intensity is not proportional to the emission cross section because the fluorescence exhibits an angular distribution and polarization that varies with the energy of the exciting electrons. Typically, a polarizer is set at the “magic angle” (54.7°) with respect to the electron beam axis to remove this polarization dependence. The literature for the derivation of the magic angle value assumes the polarizing element is perfect. An expression for the angle that accounts for the use of a partial polarizer is presented. © 2008 Optical Society of America

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1. INTRODUCTION

When a beam of particles excites a target gas and causes it to fluoresce, the cross section for excitation of a given state as a function of incident particle energy is called the optical excitation function (OEF). Typically, OEFs are measured in a cylindrical excitation geometry with the light detected at right angles to the exciting beam [1,2]. Unfortunately, the intensity of the light emitted in this direction is a function not only of the emission cross section, but also of the target alignment, i.e., the extent to which its excited electron distribution is prolate or oblate. Aligned excited states decay anisotropically, and the light emitted in any given direction is linearly polarized [3]. In reporting an OEF, it is important to remove any alignment dependence from the data so that it is proportional to the excitation cross section only. One solution is to measure the intensity in a small solid angle oriented at 54.7° with respect to the electron beam [1,4]. In the more common scenario where the light is measured perpendicular to the exciting electron beam, polarizing elements can be used to extract both the intensity and polarization. These measurements can then be used to construct the total emission cross section [1,5]. If knowledge of the polarization is not required, however, then the simple method of placing a polarizer so that its pass axis makes an angle of 54.7° to the beam is sufficient, provided the polarizer is ideal [1,4,5]. The use of a partial polarizer for this arrangement, which has apparently not been explicitly discussed in the literature, is now addressed.

2. THEORY

The fluorescence radiation from an atomic or molecular source is modeled by three dipoles oriented along the co-ordinates shown in Fig. 1 [1,4,6,7]. These radiating dipoles give rise to three intensities per unit solid angle: \( I_x \), \( I_y \), and \( I_z \) (measured perpendicular to the corresponding dipoles at some fixed radius from the origin). The electron beam is taken along the \( z \) axis. Imposing cylindrical symmetry requires \( I_x = I_y = I_z \). It is sufficient to consider only one particular value of \( \phi \), and \( \phi = \pi/2 \) is chosen for convenience. The imperfect analyzing power of a linear polarizer (pass axis oriented at an angle \( \alpha \) with respect to the \( yz \) plane) can be described by \( k \) such that the first row of its Mueller matrix is represented as [8]

\[
M_{1j} = \frac{k_1 + k_2}{2} \begin{bmatrix} k \cos(2\alpha) & k \sin(2\alpha) & 0 \end{bmatrix},
\]

where \( k_1 \) and \( k_2 \) are the maximum and minimum transmittances of the partial polarizer and

\[
k = \frac{k_1 - k_2}{k_1 + k_2}.
\]

The intensity transmitted through the polarizer for some detection angle \( \theta \) and polarizer orientation \( \alpha \) is

\[
\frac{dI(\theta, \alpha)}{d\Omega} = \frac{k_1 + k_2}{2} [I_x \sin^2(\theta) [1 + k \cos(2\alpha)] + I_y \cos^2(\theta) [1 + k \cos(2\alpha)] + I_z [1 - k \cos(2\alpha)]].
\]

Using the definition of polarization

\[
\frac{dI(\theta, \alpha)}{d\Omega} = \frac{k_1 + k_2}{2} [I_x \sin^2(\theta) [1 + k \cos(2\alpha)] + I_y \cos^2(\theta) [1 + k \cos(2\alpha)] + I_z [1 - k \cos(2\alpha)]].
\]

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it is desired that

\[ P = \frac{I_z - I_{xy}}{I_z + I_{xy}}. \]

Eq. (3) can be expressed as

\[ \frac{dI(\theta, \alpha)}{d\Omega} = \frac{(k_1 + k_2)(I_z + I_{xy})}{2}[1 + P k \cos(2\alpha) - P \cos^2(\theta)[1 + k \cos(2\alpha)]]. \]

The total intensity in the solid angle is the sum of two intensities taken with orthogonal polarizer positions, namely,

\[ \frac{dI(\theta)}{d\Omega} = \frac{dI(\theta, \alpha)}{d\Omega} + \frac{dI(\theta, \alpha + \pi/2)}{d\Omega} = (k_1 + k_2)(I_z + I_{xy})[1 - P \cos^2(\theta)]. \]

Integrating this over \( d\Omega = \sin(\theta) d\theta d\phi \) gives the total intensity

\[ I = 4\pi(k_1 + k_2)(I_z + I_{xy})(1 - P/3). \]

Using Eq. (7), Eq. (5) can be multiplied by unity in the form

\[ I = \frac{4\pi(k_1 + k_2)(I_z + I_{xy})(1 - P/3)}{4\pi(k_1 + k_2)(I_z + I_{xy})(1 - P/3)} = 1 \]

(8)

to obtain

\[ \frac{dI(\theta, \alpha)}{d\Omega} \propto I \left[ \frac{1 + P k \cos(2\alpha) - P \cos^2(\theta)[1 + k \cos(2\alpha)]}{1 - P/3} \right]. \]

(9)

Thus, for the measured intensity to be independent of \( P \), it is desired that

\[ -P/3 = P k \cos(2\alpha) - P \cos^2(\theta)[1 + k \cos(2\alpha)]. \]

(10)

If \( k = 0 \), meaning there is no polarizer, the solution of Eq. (10) is the magic angle (and its supplement) \( \theta = \cos^{-1}(\pm 1/\sqrt{3}) \). If \( 0 < k \leq 1 \) then solving Eq. (10) for \( \alpha (-\pi/2 \leq \alpha \leq \pi/2) \) in terms of \( k \) and \( \theta \) gives

\[ \alpha = \pm \frac{1}{2} \cos^{-1}\left( \frac{3 \cos^2(\theta) - 1}{3k \sin^2(\theta)} \right). \]

3. CONCLUSIONS

Equation (11) gives possible polarizer orientations \( \alpha \) as a function of \( \theta \) and \( k \). Figure 2 shows plots of \( \alpha \) versus \( k \) for various values of \( \theta \). When \( \theta \) equals the magic angle (or its supplement), Eq. (11) requires that the polarizer be set at 45°. As shown above, however, no polarizer is required at this collection angle. Note that for \( k < 1 \) this method of measuring OEFs is only valid if the instrumental polarizations of the detection elements following the polarizer are negligible. This is often a good approximation.

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