The Impact of Spectral and Spatial Exciton Distributions on Optical Emission from Thin-Film Weak-Microcavity Organic Light-Emitting Diodes

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Abstract—We present an analytical model for the optical emission produced by sources located in a thin-film weak-microcavity formation and study the effects of the ensemble spectral and spatial distribution on the device emission properties. However derived for a general stratified media configuration, the formulation results are highly applicable for the study of nanometric organic light-emitting devices. Rigorously developed into closed-form analytical expressions using the device’s thin-film weak-microcavity characteristics, they enable clear observation of the underlying physical processes that determine the emission properties of the device, as well as the impact of the exciton ensemble spectral and spatial distributions on these properties. For the sake of simplicity and clarity, we focus on a 2-D canonical configuration excited by impulsive (line) sources. Our results show that the spectral distribution of the ensemble diminishes interference effects originated in the weak microcavity formed between the substrate/air and cathode/active layer interfaces, while the spatial distribution can only impact the slow-varying component of the emission pattern, which is the consequence of the source-image interference near the highly reflecting cathode. For a typical device, the quasi-Lambertian emission pattern reported experimentally is reproduced. It should be pointed out that the incorporation of both rigorous electromagnetic analysis and the source spectral and spatial broadening effects is addressed in our report, to the best of our knowledge, for the first time. This results in a precise model capable of repeating and interpreting experimental and simulated data.

Index Terms—Electromagnetic analysis, emission pattern, excitons, microcavity, organic light-emitting diodes.

I. INTRODUCTION

O\textsuperscript{PTICAL} emission from sources embedded in thin-film stratified media has drawn major interest in the late 1970s, mainly due to the research of fluorescent entities behavior in various environments, which led to the publication of several rigorous electromagnetic analyses [1]–[3]. The problem regains interest in recent years, as the emerging field of organic light-emitting diodes (OLEDs) presents new challenges related to such formations [4]–[6]. The prospects of these devices for novel optoelectronic applications, such as thin and flexible displays, low-cost lasers, and efficient clean-energy lighting instruments [7]–[12], attract researchers to reveal the physics dominating the device and use these insights for optimizing it for the various applications. The technological effort to design durable and efficient OLEDs with controllable viewing angle [12]–[16] advances hand in hand with the ability to accurately model the emission efficiency and emission pattern of such devices, the underlying physical phenomena, and the dominant factors that determine the device’s optical properties [17]–[21].

In this paper, we present a rigorous electromagnetic approach for the OLED radiation problem capable of handling analytically the realistic scenario of an ensemble of excitons, rather than a single coherent emitter. This approach results in closed-form analytical expressions, encapsulating the effects of both the device structure, i.e., layer dimensions and material composition, and the ensemble characteristics, i.e., the spectral and spatial exciton distributions, on important optical parameters. These expressions allow identification of the dominant physical processes as well as the impact of each of them on the device’s emission pattern. For the sake of simplicity and clarity, we focus on a 2-D canonical configuration excited by impulsive (line) sources, instead of using the more realistic 3-D dipole model. However, the essence of the problem and the physical phenomena remain the same, and insight gained by the results can, in general, be applied to 3-D devices as well.

II. THEORY

A. Formulation

We consider a 2-D device with $M = N + 2$ layers, with a line source embedded at a certain plane $z = z'$, sandwiched between layers $(-1)$ and $(+1)$, as depicted in Fig. 1. The homogeneous layer formed by combining layers $(-1)$ and $(+1)$, containing the line source, is termed the active layer. Each layer is characterized by its permittivity, permeability, and conductivity marked $\varepsilon_n$, $\mu_n$, and $\sigma_n$, respectively, for the $n$th layer. Furthermore, the $r$th and $(r + 1)$th layers are separated by the plane $z = d_r$ for $n > 0$ and $z = d_{n-1}$ for $n < 0$, and we define $d_0 = z'$ and $d_{M-1} = z$. Note that $\varepsilon_{-1} = \varepsilon_1$, $\mu_{-1} = \mu_1$, and $\sigma_{-1} = \sigma_1$. For the sake of completeness, we treat here both transverse electric and transverse magnetic modes, excited via an electric line source and a magnetic line source (Fig. 1), having current magnitudes of $d_0$ and $d_{M-1}$, respectively.
Both sources are assumed to be time harmonic, with time subscripts to denote electric or magnetic cases, respectively.

B. Spatial and Spectral Distributions

Throughout the paper, we use e and m left superscripts or subscripts to denote electric or magnetic cases, respectively. Both sources are assumed to be time harmonic, with time dependence of $e^{j\omega t}$. The wave number and wave impedance of the nth layer are given as $k_n = \omega [j \mu_n \epsilon_n (1 - j \mu_n \epsilon_n / (\omega \sigma_n))]^{1/2}$ and $Z_n = \mu_n / [j \epsilon_n (1 - j \mu_n \epsilon_n / (\omega \sigma_n))]^{1/2}$, where $\epsilon$, $\gamma_n$, and $\sigma_n$ denote the dielectric constant, refraction index, and extinction coefficient, respectively. To satisfy the radiation condition we require that $\Delta [k_n] \leq 0$ leading to $\Delta [Z_n] \geq 0$. Furthermore, we define the 2-D space vector, $\vec{p} = \rho \hat{t} + \zeta \hat{\zeta}$, and $\rho$ and $\zeta$ are its transverse coordinate magnitude and direction, and $\hat{t}$ is the angle between the $z$-axis and $\vec{p}$. The transverse coordinate is different for the electric and magnetic cases due to the different symmetry they induce. In the electric line source scenario, there is no change along the $x$ direction; therefore, $\hat{t}/|\hat{t}| = 0$, $\rho' = y$, and $\zeta' = \zeta$. An analogously, for the magnetic line source scenario we have symmetry along the $y$-axis; thus, $\hat{t}/|\hat{t}| = 0$, $\rho' = x$, and $\zeta' = 1$. The source vector in both cases is $\vec{p}' = \zeta' \hat{t}'$.

C. Power Relations

Given the impulse response of the device, or Green's function, $G(\rho,t;\zeta,\tau)$, to a line source excitation at a time $\tau$ and a position $\vec{p}'$, measured at the observation point $\vec{p}$ at a time $t$, the response to an ensemble of sources, with arbitrary temporal and spatial distributions, $p(\rho')$ and $f(\zeta')$, is given by the convolution operator

$$\Theta_{int}(\rho,t) = \int d\zeta' d\rho' f(\zeta') \delta(\rho - \rho') \Theta(\rho',\rho' + \zeta';t - \tau).$$

We use the temporal Fourier transform in order to reformulate (1) into

$$G_{ax}(\rho,\omega) = \int d\zeta' d\rho' \{ p(\rho') e^{-j\omega\rho'} \} G(\rho',\rho' + \zeta';\omega).$$

where the relation between $p$ and $p$ (or $G$ and $\Theta$) is given through the Fourier transform, i.e., $p(\rho') = \int \rho' p(\rho') e^{j\omega\rho'} d\rho'$ and $p(\rho) = 1 / \omega \int p(\rho') e^{-j\omega\rho'} d\rho'$. The spectral distribution of the sources, $p(\omega)$, is related to the density of radiatively decaying states, thus resembling photoluminescence spectra [22]. The latter can be usually approximated by a Gaussian [7], [23], [24] and we define, generally

$$p(\omega) = \frac{1}{\Delta \omega} \frac{\exp \left\{ -\frac{(\omega - \omega_0)^2}{2 \Delta \omega^2} \right\}}{2 \Delta \omega^2} \text{ (3)}$$

where $\omega_0$ is the central angular frequency of the ensemble, $\Delta \omega$ is the distribution spectral width, and $\int p(\omega) d\omega = 1$. When several molecular levels contribute to the emission, the photoluminescence is more accurately modeled by a sum of such Gaussians [11], [25], and the formulation can be generalized accordingly. The spatial distribution of sources, being proportional to the variation of the recombinnation rate along the active layer, can be derived from the solution of the transport equations in the device. In most cases, this solution produces a distribution function in the form of two exponentials, decaying from their common maximum within the active layer toward the cathode and anode [26]–[29]; thus, we define

$$f(\zeta') = \frac{1}{P} \exp \left\{ -\frac{1}{W(p)} \frac{(\zeta' - \zeta_0)}{W(p)} \right\} \text{ (4)}$$

where $W(p)$ is the exponential decay width toward the cathode $\zeta_0 = 1$ and anode $\zeta_1 = -1$, $\zeta_0$ is the distribution peak location, and $W(p)$ is a normalization constant assuring that the total probability of sources to be found in the active layer is 1, or, $\int_{\zeta_1}^{\zeta_0} f(\zeta') d\zeta' = 1$. Once again, in the more peculiar forms of spatial distributions [30], the model can be readily augmented to include a sum of arbitrary exponentials as the distribution function.

The symmetry of the problem allows us to express the electric and magnetic fields by their transverse components alone. These components are related to the Green function via

$$E_\rho(\rho',\omega) = j k Z AG(\rho',\omega - \omega) - M \frac{\partial G(\rho',\omega - \omega)}{\partial \rho'} \text{ (5)}$$

$$H_\rho(\rho',\omega) = -j k ZM G(\rho',\omega - \omega) \text{ (6)}$$

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where \( S_p \) and \( M_p \) denote the Green function and transverse wave-vector, respectively. The wave equation and associated constraints for \( g(\zeta, \omega) \) are outlined in [32]-[34]. Expanding the work by Einziger et al. [34] we express the Green function in the various layers using recursive relations. For \( z > \zeta \), or \( n > 0 \)

\[
g_n(\zeta, \omega) = \frac{e^{\sqrt{n}k_\omega z}}{2\beta_N} \left[ \frac{1 - R(\zeta) e^{-2\beta_N k_\omega z}}{1 - R(\zeta) R(\zeta)} \right] \sum_{k=0}^{\infty} \left[ e^{\beta_N k_\omega z} R(\zeta) \right]^k \]

where \( g_n(\zeta, \omega) \) specifies \( g(\zeta, \omega) \) at the nth layer, \( \beta_n = \sqrt{n^2 k_\omega^2 - \xi^2} \) is the wave number in the propagation direction \( \xi \), and \( R(\zeta) \) and \( T(\zeta) \) are the total reflection and total transmission coefficients, respectively, in the forward direction, i.e., for \( n > 0 \). These coefficients are recursively defined via

\[
R(\zeta) = \left\{ \begin{array}{ll}
\Gamma(k) + (1 - \Gamma(k)) & (1 - \Gamma(k)) e^{2\beta_N k_\omega z} + e^{-2\beta_N k_\omega z} \\
1 + \Gamma(k) & e^{2\beta_N k_\omega z} + e^{-2\beta_N k_\omega z} \end{array} \right.
\]

\[
T(\zeta) = \left\{ \begin{array}{ll}
\Gamma(k) + (1 - \Gamma(k)) & (1 - \Gamma(k)) e^{2\beta_N k_\omega z} - e^{-2\beta_N k_\omega z} \\
1 + \Gamma(k) & e^{2\beta_N k_\omega z} - e^{-2\beta_N k_\omega z} \end{array} \right.
\]

where \( \Gamma(k) \) is the forward local reflection coefficient of the nth interface, \( n > 0 \), given by the Fresnel formula, \( \Gamma_n = \frac{1 - t_n}{1 + t_n} \). We used the definition of the generalized impedance ratio, \( \omega_N = (\omega_N + \omega_N)/(1 + \omega_N) \). The analogous expressions for the reflection and transmission coefficients in the reversed direction, \( R(\zeta) \) and \( T(\zeta) \), etc., for \( n < 0 \), can be readily derived in a similar manner [35], using the transformations \( (n - 1) \leftrightarrow (n + 1) \) and \( j \leftrightarrow (-j) \). The recursion stop conditions are \( R_{N+1} = R_{N-1} = 0 \). Substituting (6)–(8) into (5), and taking only the saddle point contribution to the plane wave spectral integral into account [33] we arrive at the following expression for the emission pattern:

\[
S_p(\theta, \phi) \sim P_{\zeta} e^{\delta j f(\zeta)} \sum_{\zeta} \int_{-\Delta \omega} d\omega \left\{ \frac{2 M_{\zeta} \mathbf{g}(\zeta, \omega)}{\sqrt{\beta_N^2 - \xi^2}} \right\}^2
\]

where \( P_{\zeta} = k_N Z_N \mathbf{g}(\zeta, \omega) / 16 \) and \( M_{\zeta} = k_N V_\zeta \mathbf{g}(\zeta, \omega) / 16 \) denote the radiation power of electric and magnetic line sources in an unbounded homogeneous medium of the nth layer material, respectively. Following previous work [36], we express the 1-D Green function in the observation region, \( n = N + 1 \), for \( N \geq 1 \), as a sum of multiple reflections between the layers’ boundaries

\[
g_{N+1}(\zeta, \omega) = \sum_{k=0}^{\infty} \left[ e^{\beta_N k_\omega z} R_N \right] ^k \sum_{l=0}^{\infty} \left[ e^{-\beta_N k_\omega z} T_N \right] ^l
\]

\[
\sum_{k=0}^{\infty} \sum_{l=0}^{\infty} e^{-2\beta_N k_\omega z} \left( 1 + e^{2\beta_N k_\omega z} \right)
\]

D. Spectral Distribution Effect

Assuming that in the relevant spectral interval of (3), \( \omega_N \ll \Delta \omega_N \), the optical parameters of the media, i.e., \( n_0 \) and \( k_0 \), do not vary significantly with \( \omega \), we get that after the substitution \( k_N \sim \sqrt{\omega_N} \) of (10), all \( \beta_N \) are proportional to \( \omega_N \) and all \( \Gamma_N \) and \( \Gamma_0 \) are independent of the field’s angular frequency. Incorporating (11) into (10), the latter can be reformulated as a sum of multiple reflection terms, and the spectral and spatial integrals can be solved, term-by-term. Each term takes the form of a multiple reflection combination, consisting of a reflection coefficient product, \( a_{\phi}(\theta) \), independent of \( \omega_N \), and a phase factor, \( e^{-2\phi_{\omega_N}(\theta)} \equiv e^{-2\phi_{\omega_N}(\theta)} \), accumulating a linear phase relative to the propagation induced by the multiple reflections defined by \( a_{\phi}(\theta) \). The parameters \( I_1(\theta) \) and \( \tau_1(\theta) \) define the optical propagation length and propagation time, respectively, within the device, and are also independent of the angular frequency. The spectral integral for a single term can therefore be solved analytically, leading to

\[
\int_{-\Delta \omega} d\omega e^{-2\phi_{\omega_N}(\theta)} = \sum_{\phi_{\omega_N}(\theta)} \phi_{\omega_N}(\theta) e^{-2\phi_{\omega_N}(\theta)} = \int_{-\Delta \omega} d\omega e^{-2\phi_{\omega_N}(\theta)}
\]
where the coherence length for the ensemble, \( L_c = \frac{\lambda}{\delta \lambda} = \lambda^2/(2\Delta \lambda) \), is defined as the optical length in air until which two photons generated by the ensemble at the same moment with the same initial phase can be still considered coherent [37], i.e., their frequency difference does not induce a significant phase difference upon propagation distance \( L_c \). The use of the term “coherence length” in this context does not imply a constant phase difference from different excitons (their emission was already shown to be good approximation is (8)), as all other factors are independent of the source distribution cases, where \( z \) is the effective wavelength for the \( r \)-th term in (14) experiences an additional attenuation due to the spectral distribution of the sources, and the corrected spatial broadening factor, \( F \), must be used.

**F. Closed-Form Solution for Prototype Device**

In order to demonstrate the impact of the last two results, i.e., the effect of spectral and spatial source distributions on the optical emission from thin-film weak-microcavity formations, we apply them to a prototype device. A basic configuration of an elementary device, specified in Table I and depicted in Fig. 2, is selected [7]. The optical emission from thin-film weak-microcavity formations, i.e., the effect of spectral and spatial source distributions on the location-dependent terms will be introduced in Section II-F later on), and the effect of the spatial broadening on the location-dependent terms cannot be totally neglected. In those cases, we must execute the spatial integral as well, leading to

\[
\int_{z}^{z'} d\zeta f(\zeta) = \sum_{r=1,0.1}^{d_{r,1}} \sum_{u=1,0.1}^{W_{r} \cdot W_{u}} \left[ \frac{1 - e^{-(\zeta_{r} - \zeta_{u})/W_{r,1}}}{\zeta_{r} - \zeta_{u}} \right] \]

which is an accurate expression if \( \zeta_{r} = d/2 \). For other values of \( \zeta_{r} \), the spatial broadening factor lies between these two limiting cases.

For exciton ensembles located far from the active layer boundary at \( d_1 \), the phase term \( (\zeta_{r} - d_{r,1}) F_{1}(\theta, \omega) \) becomes comparable with the typical coherence length (typical values will be introduced in Section II-F later on), and the effect of the spatial broadening on the location-dependent terms cannot be totally neglected. In those cases, we must execute the spatial integral as well, leading to

\[
\int_{z}^{z'} d\zeta f(\zeta) = \sum_{r=1,0.1}^{d_{r,1}} \sum_{u=1,0.1}^{W_{r} \cdot W_{u}} \left[ \frac{1 - e^{-(\zeta_{r} - \zeta_{u})/W_{r,1}}}{\zeta_{r} - \zeta_{u}} \right] \]

where the Gaussian decay varies much slower with \( \zeta \) than the spatial distribution factor, and thus can be removed from the integrand for the spatial integration. For typical exciton spatial distributions and coherence lengths the Gaussian factor remains close to unity; however, in some cases, as discussed above, the \( r \)-th term in (14) experiences an additional attenuation due to the spectral distribution of the sources, and the corrected spatial broadening factor, \( F \), must be used.

**E. Spatial Distribution Effect**

Solving the spatial integral involves only the first two factors in (8), as all other factors are independent of the source location, \( \zeta' \). Referring to the relevant form of these factors in (10),

\[
\int_{z}^{z'} d\zeta f(\zeta) = \sum_{r=1,0.1}^{d_{r,1}} \sum_{u=1,0.1}^{W_{r} \cdot W_{u}} \left[ \frac{1 - e^{-(\zeta_{r} - \zeta_{u})/W_{r,1}}}{\zeta_{r} - \zeta_{u}} \right] \]

where \( L_i(\theta, \omega) \) is the effective wavelength for the \( r \)-th term, and \( F_0(\zeta_{r}, \zeta_{u}) \) is the spatial broadening factor for the \( r \)-th term. It is readily observed that if the spatial widths \( W_{r,1} \) are very small with respect to the active layer dimensions, then \( F_0(\zeta_{r}, \zeta_{u}) \) approaches 1, and \( f(\zeta) \) takes the form of a delta function, as expected. On the other hand, if the spatial broadening is substantial, \( F_0(\zeta_{r}, \zeta_{u}) / F \) decreases the weight of the spatial term as the spatial widths \( W_{r,1} \) increase. This can be inferred from the approximations we present for the symmetric distribution cases, where \( W_1 = W_{r,1} = W \). If the ensemble center is close to the active layer boundaries, \( \zeta_{r} \rightarrow d_{r,1} \), a good approximation is

\[
F_0(\zeta_{r}, \zeta_{u}) \approx \frac{1 - j(2W/L_c)}{1 + j(2W/L_c)} \]

whereas when the excitons are concentrated around the middle of the active layer, \( \zeta_{r} \rightarrow (d_{r,1} - d)/2 = d/2 \), the spatial broadening factor is given by

\[
F \frac{F_0(\zeta_{r}, \zeta_{u})}{\zeta_{r} - \zeta_{u}} \approx 1 \pm j(2W/L_c) \frac{2W/L_c}{1 + j(2W/L_c)} \]

(16)

where the coherence length for the ensemble, \( L_c = \frac{\lambda}{\Delta \lambda} = \lambda^2/(2\Delta \lambda) \), is defined as the optical length in air until which two photons generated by the ensemble at the same moment with the same initial phase can be still considered coherent [37], i.e., their frequency difference does not induce a significant phase difference upon propagation distance \( L_c \). The use of the term “coherence length” in this context does not imply a constant phase difference from different excitons (their emission was already shown to be good approximation is (8)), as all other factors are independent of the source distribution cases, where \( z \) is the effective wavelength for the \( r \)-th term in (14) experiences an additional attenuation due to the spectral distribution of the sources, and the corrected spatial broadening factor, \( F \), must be used.
Third, the typical emission spectral width of these devices [7], substrate thickness is greater by several orders of magnitude, i.e., they are considered thin-film layers, whereas the glass are comparable with the emission wavelength in the media, Table I.

In this section, we will present an analytical closed form we can readily identify the dominant spatial and spectral source distributions is mediated by cathode/polymer and glass/air interfaces. The impact of the positions with respect to the cathode; the direct ray contribution (21), which vanishes due to the large substrate dimensions and transmission through layers; and the weak-microcavity effects (22), which consist of multiple reflections between the cathode/polymer and glass/air interfaces. The impact of the spatial and spectral source distributions is mediated by the spatial broadening factor, $F(k) = (1/N_s) (1/v_F)$, multiplying the image-source interference term in (20), and the absence of the weak-microcavity cross-terms from (22), which vanishes due to the large substrate dimensions with respect to realistic coherence lengths. This result of the rigorous derivation is capable of reproducing the measured emission patterns as well as evaluating the quality and validity of heuristic approaches, such as semi-infinite glass (20), perturbed glass thickness [22], or ray-optics [17], [18].

III. RESULTS AND DISCUSSION

A. Spectral Distribution Effect

Emission patterns of prototype BE-OLEDs with varying either spectral distribution widths or glass thicknesses are depicted in Figs. 3 and 4, respectively. In both figures the spatial distribution is defined as the delta function, $f(\nu) =

### Table I

<table>
<thead>
<tr>
<th>$\nu$</th>
<th>Layer Material</th>
<th>$\nu_0$</th>
<th>$\nu_0\nu_0$</th>
<th>$\nu_0\nu_0\nu_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.25</td>
<td>Glass</td>
<td>1.5</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0.125</td>
<td>Air</td>
<td>1.0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

*Fig. 2. Physical configuration of the prototype BE-OLED specified in Table 1.*
In order to observe the effect of the spatial distribution on the emission pattern we must choose the center of the distribution to be in a location where the image-source interference is significant. Thus, in Figs. 5 and 6 we plot the emission patterns for a source distribution with maximum at \( z_0 = 140 \text{nm} \), and vary the distribution widths from \( W_{-1} = W_1 = 1 \text{nm} \) to \( W_{-1} = W_1 = 100 \text{nm} \). In order to stress the different impacts of the spatial and spectral broadening, Fig. 5 is plotted for the prototype device taking the coherence length to be infinite, whereas the same plots are presented in Fig. 6 with a realistic coherence length of \( L_c = 1 \mu\text{m} \). As discussed above, for small spatial distribution widths, the distribution function approaches the delta function; therefore, the response for these parameter values in Fig. 5 is the same as for a single coherent source.
When the distribution width increases, the original image-source interference pattern disappears, as the interference term is diminished according to (14). Similar attenuation of image-source interference patterns due to exciton spatial distribution broadening can be observed in the results of Savaidis and Stathopoulos [21], where the authors present simulated OLED electroluminescence spectra, which also usually contain extreme shifts in the image-source interference extrema, due to the different metal/polymer reflection coefficient phase. Nevertheless, as formulated in (17) and discussed therein.

It is worth noting that emission patterns associated with the commonly used aluminum cathode will experience minor shifts in the image-source interference extrema, due to the different metal/polymer reflection coefficient phase. Nevertheless, the formulation results as well as the essence of the physical processes remain the same.

IV. CONCLUSION

A complete analysis for the optical emission from 2-D thin-film weak-microcavity formations, incorporating spatial and spectral source distributions effects, has been presented and verified through numerical simulations for a basic BE-OLED configuration. The results imply that realistic values of spectral and spatial distribution widths can explain the transformation between emission patterns typical to a single coherent source and the experimentally observed quasi-Lambertian patterns. Moreover, the closed-form expressions derived for a prototype BE-OLED reveal that the exciton ensemble spectral broadening is responsible for the vanishing of the fast-varying component of the emission pattern, related to the weak-microcavity interference effects, whereas the effect of the spatial distribution is limited to the slow-varying component of the emission pattern, resulting from the image-source interference induced by the presence of the metallic cathode. These resultant analytical expressions preserve the physical intuition of the device optics and allow for an efficient implementation and design. Furthermore, as demonstrated for the prototype device, they establish a clear and simple relation between the device structure, the ensemble statistics, and the emission pattern.

REFERENCES


