Rigorous optical modeling of multilayer organic light-emitting diode devices

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We present an exact classical solution to the problem of dipole emission in a planar multilayer light-emitting device. The inputs to the model are the photoluminescence and quantum yield of the emitter material, and the device layer thicknesses and indices of refraction. The results of the model are applied to predicting the radiant intensity of organic light-emitting diodes as a function of varying device layer thickness. It is shown that the predicted radiances are in excellent agreement with the data. We also present results for the Poynting power distribution from a randomly aligned dipole for positions both internal and external to the diodes. © 2001 American Institute of Physics. [DOI: 10.1063/1.1356453]

Recently there has been increasing interest in computing the optical emission properties of organic light-emitting diodes (OLEDs) because of their application as lightweight, low-cost display sources. In order to optimize the spectral output of OLED devices, it is useful to have an optical model that can calculate both the color and intensity of the radiated emission as a function of variations in the device design. In addition, since a large fraction of the emitted light does not exit the device, it is also helpful to have an optical model that can compute the wave intensity inside the device.

The optical models to date can be split up into three general schemes: (1) use a radiative transfer approach, which allows one to compute only the color; (2) use results from the model of Chance et al. which enables one to compute accurately the intensity, but only for simple device structures; and (3) use Fermi’s golden rule to compute both the radiative and internal field intensities; however, the validity of that approach is limited to lossless devices. In this letter we discuss a formalism that enables one to compute all three desired quantities: color, radiative intensity, and the internal power distribution. The methodology is based on a Hertzian vector approach to the inhomogeneous vector wave equation

\[
(\nabla^2 + k^2)\Pi^{(e)} = -\mu \mathbf{P}_0 / \varepsilon_0 n^2,
\]

where \(\Pi^{(e)}\) is the electric Hertzian vector, \(\mathbf{P}_0\) is the electric polarization vector due to the oscillating dipoles, \(n\) is the wavelength-dependent complex refractive index (measured for each layer in the device by spectroscopic ellipsometry), and \(k = 2\pi n/\lambda\). Equation (1) is valid for the case where there are no magnetic dipoles present in the media. The electric polarization can be written in terms of the dipole moment, \(\mathbf{p}\), as \(\mathbf{P}_0 = \mathbf{p} \delta(r - r')\).

The solution to Eq. (1) splits up into two cases: the dipole oriented parallel to the layers (horizontal dipole), and the dipole oriented perpendicular to the layers (vertical dipole). It will be assumed here that the OLED layers are grown in the \(z\) direction (vertical). As a result of the cylindrical symmetry of the problem for the vertical-dipole case, the formalism for the horizontal-dipole case is slightly more involved and is discussed elsewhere. The formalism for the \(j\)th layer, \(\Pi_{j-}\), can be expanded as (the subscript \(z\) denotes that \(\Pi^{(e)}\) is directed in the \(z\) direction)

\[
\Pi_{j-} = -\frac{i\rho}{4\pi \varepsilon_0 n_e^2} \int_0^\infty \left[ T_{j \alpha}(\alpha) z e^{i\gamma(z-z_f)} \right. \\
\left. + T_{j \alpha}(\alpha) e^{-i\gamma(z-z_f)} J_0(\rho \alpha) \, d\alpha, \right.
\]

where the \(T_j\) are expansion coefficients which are determined by application of the boundary conditions, \(\gamma^2 = k^2 - \alpha^2\), \(n_e\) is the index of refraction of the emission layer, \(z_j\) is the lower boundary of the \(j\)th layer, \(J_0\) is a Bessel function of the first kind, and \(\rho\) is the cylindrical radial coordinate. Equation (2) applies to nonemission layers; for the emission layer a dipole source term must be added to \(\Pi_{j-}\). Upon differentiating Eq. (2) with respect to \(\rho\) and \(z\), analytical expressions for the \(\mathbf{E}\) and \(\mathbf{H}\) fields are obtained. Since the integrals cannot be performed in closed form for the general case of an \(n\)-layer device, they need to be evaluated numerically using an adaptive quadrature routine.

In the far-field \(H\) is proportional to \(E\); therefore, the Poynting power scales as \(|E|^2\). In order to compute the
wavelength-dependent radiated intensity, \( I_r(\lambda) \), it is also necessary to have an expression for the total power, \( P_{\text{tot}} \), emitted by the dipole. Typically, \( P_{\text{tot}} \) has been approximated by integrating over the power radiated into the far field by the device.\(^7\) Using our formalism the \( \mathbf{E} \) and \( \mathbf{H} \) fields are known everywhere, which enables one to evaluate \( P_{\text{tot}} \) exactly by integrating the power over the surface of a small sphere that surrounds the dipole. As a result, \( I_r(\lambda) \) is given by

\[
I_r(\lambda) = \frac{|\mathbf{E}|^2 S_0(\lambda)}{P_{\text{tot}} + P_d/q},
\]

where \(|\mathbf{E}|^2\) is computed as a function of wavelength, \( S_0(\lambda) \) is the wavelength-dependent dipole oscillator strength, \( P_{\text{tot}} \) is the total power minus the direct contribution, \( P_d \), from the isolated dipole, \( q = P_d/P_{\text{tot}} \) is the quantum yield of the isolated dipole, and \( P_{d,\text{tot}} \) is the inverse lifetime of the isolated dipole. The magnitude of \( q \) is one of the two adjustable parameters in the model, and we used the typical value of 0.8.\(^6\) The other unknown in Eq. (3) is \( S_0 \); it was assumed to be proportional to the measured photoluminescence (PL) response of the emitter material. To be more specific, we had to rigidly blueshift the PL response by 4 nm (this is the other adjustable parameter in the model) in order to get good color predictions. It should be noted that the results for \(|\mathbf{E}|^2\) are averaged over a few thicknesses of the glass since the glass thickness of 0.7 mm is much larger than the coherence length of the emitted light. In addition, \(|\mathbf{E}|^2\) is also averaged over the position of the dipole within the recombination zone; this zone is taken to be 100 Å thick and borders the hole transport layer (HTL)-emitter layer heterojunction.\(^12\)

Figure 1 compares the calculated radiances, Eq. (3), with the measured data for OLED devices where the HTL thickness varies from 800 to 2400 Å (only 2 of the thicknesses are shown in the figure). The devices were grown by high vacuum \((\sim 10^{-6}\) Torr) thermal evaporation onto the surface of an indium tin oxide (ITO) coated glass substrate. Following the ITO (850 Å thick) was grown the HTL composed of 4,4’-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB), 300 Å of aluminum tris(8-hydroxyquinoline) (Alq) doped with 0.5% of Coumarin 545T (C545T), 500 Å of undoped Alq in the electron transport layer (ETL), and lastly 2000 Å of MgAg at a Mg–Ag volume ratio of 10:1. All of the device measurements were performed at a constant current of 20 mA/cm\(^2\) and the radiance data was collected in the normal viewing direction. Since OLEDs are electro-optical devices, predicting the absolute intensity would require linking a transport model to our optical model. As a result, the calculations are scaled by a constant such that the predictions for the NPB thickness of 1200 Å (not shown) have the same peak value as the corresponding data. As can be seen from the figure, the model predicts the data very well (both color and intensity). It is interesting to note that the peak intensity of the data varies by approximately 40% over the thickness range, while the calculated total dipole power, \( P_{\text{tot}} \), only varies by 3%. Consequently, varying the NPB thickness has only a minor influence on the magnitude of the total dipole emission, while having a noticeable impact on the angular emission pattern of the radiation. It should be noted that making the approximation that \( P_{\text{tot}} \) can be calculated by integrating over the far-field distribution\(^2\) would result in \( P_{\text{tot}} \) varying by 25% over the thickness series.

Figure 2 shows the effect of varying the ETL (Alq) thickness between 75 and 1800 Å. The other device layer thicknesses were 850 Å of ITO, 1500 Å of NPB, and 300 Å of Alq doped with 0.5% of C545T. Again the predictions are in excellent agreement with the data. While the total dipole emission is fairly constant for the HTL thickness series, it varies by a factor of 1.8 for the ETL thickness series. This occurs because nonradiative transfer of energy from the dipole to the metal’s surface plasmons\(^6\) is a dominant recombination mechanism for emitters close to the metal’s surface. We have determined that the surface-plasmon quenching affects dipoles within \(\sim 1175\) Å of the MgAg surface. Referring back to Eq. (3), the resulting radiances are not only influenced by the varying dipole lifetime, but also by the oscillating angular emission pattern (due to optical interference effects) which has maxima at ETL thicknesses of \(\sim 400\) and 1900 Å and a minimum at \(\sim 1050\) Å. The convolution of these two effects results in the peak intensity being stronger at the second maximum (1900 Å) due to the lack of plasmon quenching at that distance from the metal. Previously, this finding was discussed by Saito et al.\(^1\)

Figure 3 shows the Poynting power contour plot for a...
randomly aligned dipole in the emission layer. For this calculation the thickness of the device layers are 1 μm of glass, 850 Å of ITO, 1500 Å of NPB, 300 Å of Alq doped with 0.5% of C545T, and 450 Å of undoped Alq as the ETL. The figure also contains arrows showing the direction of the Poynting power. Both the contours and the arrows are on a log 10 scale. The vertical lines separate the various device layers, which starting at the left are: ETL, doped Alq (emitter), HTL, ITO, and glass.

In conclusion, we have presented an exact classical solution to the problem of exciton emission in planar OLED devices. Comparisons with experimental radiance data have been made as a function of varying layer thickness. The model is shown to predict accurately the data which suggest that at constant current the variation in electroluminescence caused by modifications of the layer thicknesses can be completely accounted for by optical effects. In addition, we have presented results for the internal field distributions that should be beneficial for improving the light extraction efficiency. Even though the calculations have been geared toward organic LED devices, the formalism can be equally applied to inorganic LEDs.13

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