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# Energy transfer between host and dopant molecules in blue organic light-emitting devices

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Blue organic light-emitting devices were fabricated with an activator of 4,4'-bis(9-ethyl-3-carbazovinylen)-1,1'-biphenyl doped into the various host materials such as N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1-biphenyl-4,4'-diamine; 4,4'-bis(2,2'-diphenylynyl)-1,1'-biphenyl; 2-methyl-9,10-di(2-naphthyl) anthracene; and 1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene to investigate optical properties of blue light emission in the host-dopant system. By spectroscopic analysis based on multi-peak fits to the emission spectra, we found that energy transfer between the host and dopant molecules have a strong correlation with key features; current density, luminous efficiency, and color index for the devices. Among the present dopant-host systems, the TPBi molecule was found to be the best molecule as a host material for our devices. In contrast, the DPVBi host induced a complex excimer (electromer) leading to a shoulder spectrum with a longer wavelength emission. It was found that the electromer significantly affects the optical and electrical properties of the device. © 2011 American Institute of Physics. [doi:10.1063/1.3614493]

## I. INTRODUCTION

Organic light-emitting device (OLED) is one of the most promising display and lighting technologies owing to its color gamut.<sup>1</sup> Since Tang and van Slyke<sup>2</sup> developed multi-layered thin film OLED, its application has been widely extended from a display to a diffuse source of light.<sup>3</sup> Research on high-efficiency OLED, however, has been made mainly via experimental trial and error methods as changing layer materials and device structures. Eventually, such efforts to improve OLED efficiency have resulted in complicated multi-layer structures along with high cost and time consumption. Prior to consideration of specific experimental trial, it is useful to understand how emission spectra are affected by physical processes occurring between molecules in an emissive layer.<sup>4–13</sup> It should be emphasized that the electroluminescence (EL) characteristics of OLED can be explained in terms of the properties of individual, isolated organic molecules in the emissive layer. In many organic molecules, photons can excite electrons from the filled valence bands (occupied molecular orbitals) to the empty conduction bands (unoccupied molecular orbitals) and also interact with molecular vibrations and electrons of neighboring molecules. Therefore, the molecular energy transfer within the dopant-host system of OLED plays an essential role in determining emitting efficiency. It is well known that the light-emitting efficiency is proportional to the spectral overlap of light absorption and output emission between the dopant and host molecules in OLED.<sup>14–22</sup> To understand quantum mechanism of the energy transfer responsible for light-emitting in organic molecules, it is essential to investigate not only the electronic energy gap  $E_g$ , but also the structure of the energy levels of the molecules concerned. In fact,

by using a spectroscopic analysis for the emission spectrum, we can obtain useful information about the energy transfer in determining optical properties of the dopant and host materials and also on the physical background in explaining the molecular energy level structure. Until now, however, studies of the molecular energy level structures and the associated physical properties for high-efficiency OLEDs have rarely been made.

In this paper, we present a detailed spectroscopic study of the structure of the molecular energy levels and discuss the emission properties in terms of the energy transfer mechanism in the dopant-host system for OLEDs. The fabricated OLEDs have a multilayer structure with a blue activator of 4,4'-bis(9-ethyl-3-carbazovinylen)-1,1'-biphenyl (BCzVBi) doped into the various host materials such as N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1-biphenyl-4,4'-diamine (NPB), 4,4'-bis(2,2'-diphenylynyl)-1,1'-biphenyl (DPVBi), 2-methyl-9,10-di(2-naphthyl) anthracene (MADN), and 1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBi). The present spectroscopic study on the EL spectra of the devices plays a key role in guiding data analysis as well as in finding materials suitable for creating high-efficiency OLEDs.

## II. EXPERIMENTAL PROCEDURE

Figure 1 shows the device structure of the blue OLED. The emitting layer consists of BCzVBi doped into the host materials such as NPB, DPVBi, MADN, and TPBi, which are denoted devices A, B, C, and D, respectively. Here the molecule of BCzVBi, doped wt5% in the host material, acts as an activator with luminescence occurring in the blue region. The structures were fabricated on indium-tin-oxide (ITO) glass substrates with active areas of 4 cm<sup>2</sup>. After oxygen plasma treatment to clean up the surface of the substrates, they were loaded into the high vacuum thermal evaporator for thin film deposition of organic layers and metal electrodes. Finally the

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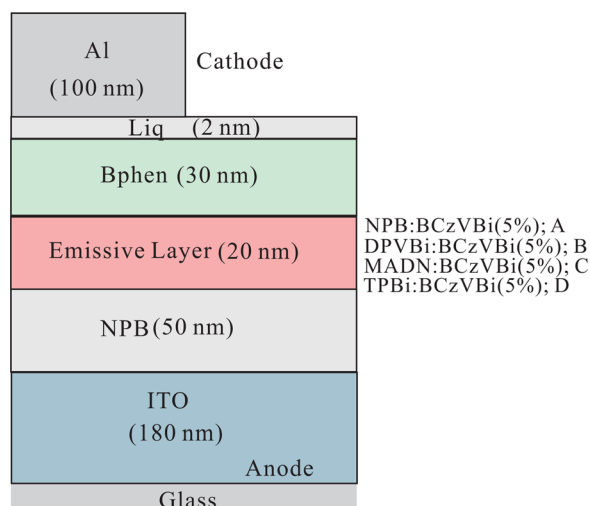


FIG. 1. (Color online) Structure of blue OLED devices A, B, C, and D.

devices were sealed by a cover film through ultraviolet exposure encapsulation method in the glovebox. NPB, 4,7-diphenyl-1,10-phenanthroline (BPhen) and lithium quinolate (Liq) were used as a hole transporting layer, an electron transporting layer (also a hole blocking layer), and an electron injecting layer, respectively. The current density versus applied bias voltages, luminance versus current density, and luminous efficiency versus current density were measured with a Keithley 236 current source measure unit, a CHROMA METER CS 100-A, and a JBS OLED analysis system IVL-2000.

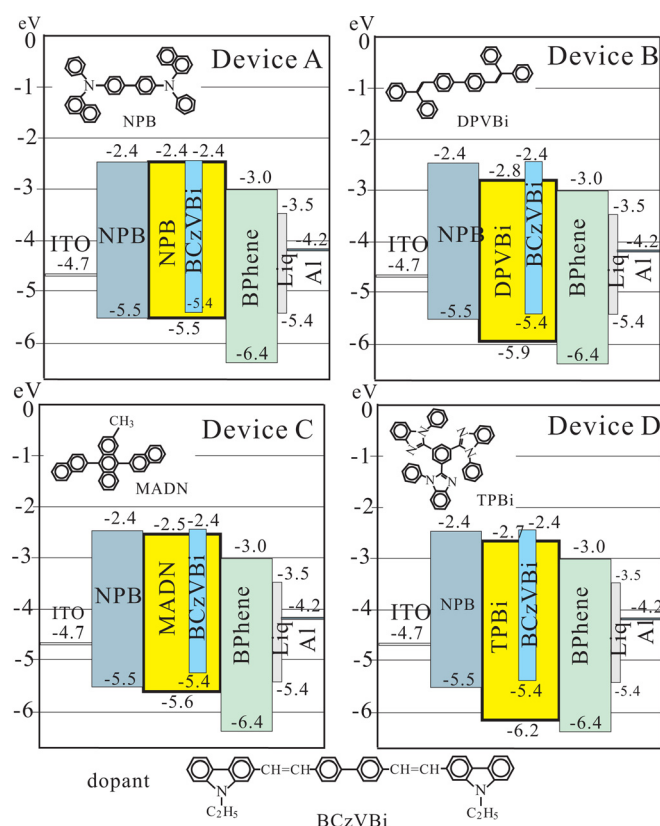


FIG. 2. (Color online) Energy level diagrams and molecular structures of organic materials with various host and dopant combinations.

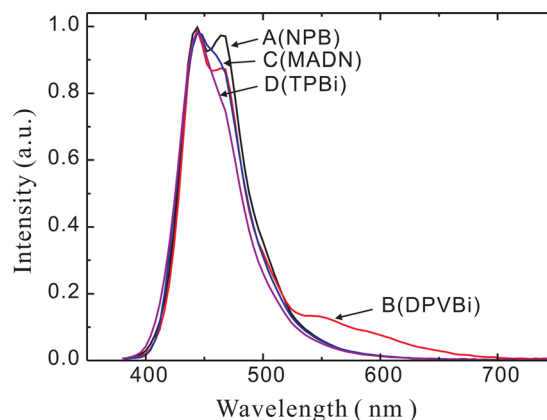


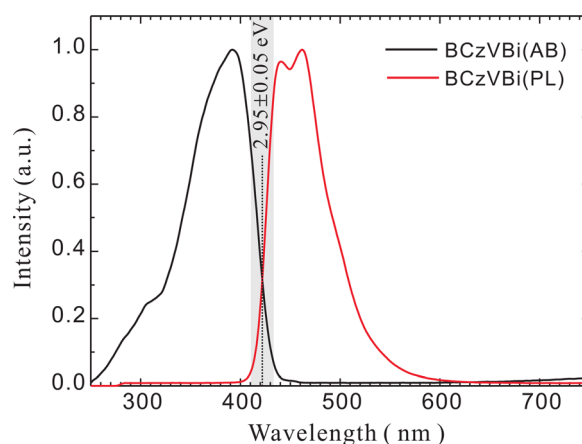
FIG. 3. (Color online) Normalized electroluminescence spectra from the present devices A, B, C, and D. See text for more details.

Energy level diagrams and molecular structures of the host and dopant materials are illustrated in Fig. 2.

### III. RESULTS AND DISCUSSIONS

#### A. Characteristics of electroluminescence spectra

Figure 3 shows the electroluminescence (EL) spectra from the present OLEDs with BCzVBi as a blue activator doped into the host materials of NPB, DPVBi, MADN, and TPBi, respectively. It should be noted that no EL originating from the host molecules was observed. There are two distinctive features in the EL spectra: One is the intensity variation of the second peak at 480 nm corresponding to the different host materials; the other is the presence of a shoulder emission in the device D with the host of DPVBi. As the starting point for spectroscopic analysis, we compared the absorption spectrum of the dopant with the output spectrum due to photoluminescence (PL). Figure 4 illustrates the light absorption and the PL emission spectra of the dopant BCzVBi after ultraviolet-visible light irradiation. From the absorption-emission edge, we estimate the apparent optical bandgap  $E_g$  (HOMO-LUMO gap) to be  $2.95 \pm 0.05$  eV. As evident in Fig. 4, the PL spectrum has two peaks at 440 nm and 460 nm (we called it the second peak) among which the second peak has higher intensity than

FIG. 4. (Color online) Normalized photo-absorption (left) and photoluminescence (right) spectra of BCzVBi in chloroform solvent. Here  $2.95 \pm 0.05$  eV corresponds to the optical bandgap energy.

the first one. By comparing it with the EL spectra as shown in Fig. 3, we recognize that the second peak intensity is strongly dependent on the host materials. In other words, this feature indicates that the corresponding second electronic transition in BCzVBi molecule is more sensitive to energy transfer through inter-molecular interactions between the dopant and the host molecules.

Figure 5 displays the ultraviolet-visible light absorption spectra of BCzVBi and the PL emission spectra of the host materials. According to the theory of energy transfer for a donor-acceptor system, the transfer probability is proportional to the spectral overlap of an absorption spectrum of the acceptor and an emission spectrum of the donor.<sup>14,15</sup> The transfer probability rate from a donor to an acceptor in this theory can be given by the Fermi golden rule expression:

$$\Gamma_{D \rightarrow A} = \frac{2\pi}{\hbar} \sum_{ij} |\langle i | U_{int} | j \rangle|^2 \delta(\epsilon_i - \epsilon_j).$$

Here  $\langle i | U_{int} | j \rangle$  is the matrix element of the electromagnetic coupling between the states and  $|i\rangle$  is the initial state, where the donor is in an excited state and the acceptor is in the ground state, and  $|j\rangle$  is the final state where the excitation was transferred to the acceptor at an excited state. Dirac delta function  $\delta(\epsilon_i - \epsilon_j)$  ensures conservation of energy between initial and final states. In the present host-dopant system, the interaction potential is given by the following dipole-dipole coupling expression

$$U_{int}^{D \rightarrow A} = \frac{1}{4\pi\epsilon} \frac{(\mathbf{p} \cdot \mathbf{p}') - 3(\mathbf{p} \cdot \hat{\mathbf{r}})(\mathbf{p}' \cdot \hat{\mathbf{r}})}{r^3},$$

where  $\mathbf{p}(\mathbf{p}')$  is a dipole moment of the donor (acceptor) and  $\hat{\mathbf{r}}$  is a unit vector of the separation  $r$  between the donor and the acceptor. Notice that  $\epsilon (= \epsilon_0 \epsilon_r)$  is the electric permittivity in a medium, where relative permittivity (or dielectric constant)  $\epsilon_r$  is dependent on optical frequencies. The final expression for the above resonance energy transfer rate is given in terms of spectral overlap integrals

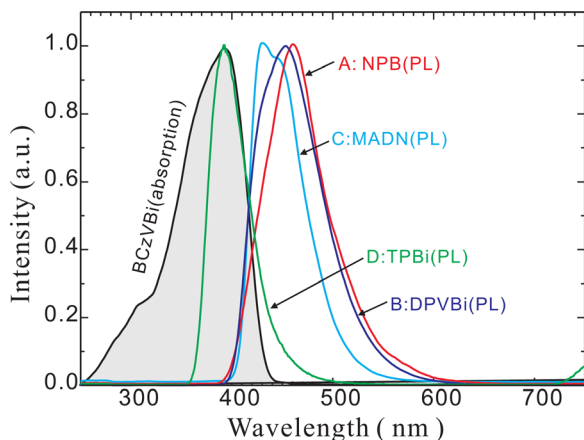


FIG. 5. (Color online) Comparison of the absorption spectrum of BCzVBi with the photoluminescence spectra of the host materials of NPB, DPVBi, MADN, and TPBi, respectively.

$$\Gamma_{D \rightarrow A}^{d \rightarrow d} = \frac{1}{12\pi\hbar\epsilon^2 r^6} \int D_{dip}(\epsilon) A_{dip}(\epsilon) d\epsilon,$$

where  $D_{dip}$  and  $A_{dip}$  are the emission and absorption spectral functions, respectively. The details are given in Ref. 14. Thus, the EL efficiency of OLEDs should be dependent on the spectral overlap between the PL peak of the host as an electron donor and the absorption peak of the dopant as an electron acceptor. It is worth emphasizing that the theory of energy transfer is based on the concept of the host as an oscillating dipole, which can exchange energy with another dipole of the dopant with a similar frequency. Among the present host materials, the PL peak of TPBi provides the largest overlap that is estimated to be about 90% of its total area. The spectral overlaps of the other host materials, however, are much smaller than that of TPBi, which is almost equal in area to each other. Because of the larger spectral overlap, the molecular interaction between the dopant of BCzVBi and the host of TPBi is greatly enhanced and thus their energy transfer is expected to be much larger than the other dopant-host systems. Furthermore, from this simple spectral overlap analysis, we find that the energy transfer is directly proportional to the energy gap difference between the host and dopant materials. Notice that NPB, MADN, and DPVBi molecules are nearly identical in the electron gap energy,  $E_g \simeq 3.1$  eV, which is only 0.1 eV higher than that of the dopant BCzVBi while TPBi has  $E_g \simeq 3.5$  eV as shown in Fig. 2.

Next we focus on the relationship between the intensity of energy transfer and the variance of the second EL peak of BCzVBi appearing in each device. For obtaining the relative intensity of the peaks, we analyzed the EL spectra with a multi-peak fit by assuming that a peak spectrum follows a Gaussian distribution. Figure 6 represents a result of the multi-Gaussian fits to the EL spectra of the devices. The multi-curve fits based on the Gaussian distribution allow the determination of the peak positions; namely, the transition energies and the relative yield for each of the two peaks. It should be noted that the spectrum of device B has a shoulder at longer wavelengths. We will discuss this point later. The

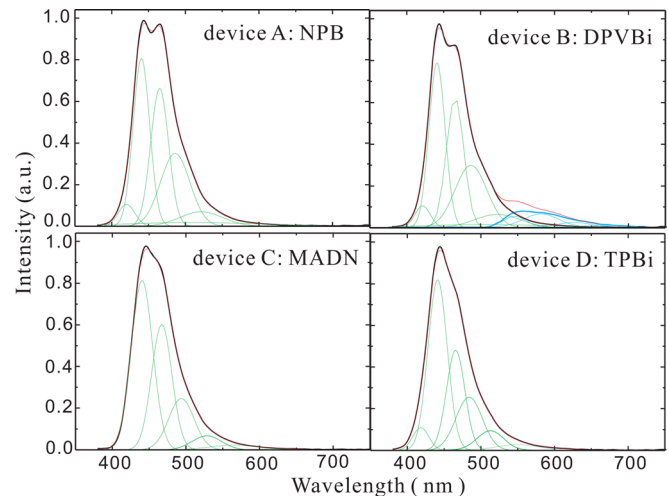


FIG. 6. (Color online) Multi-peak Gaussian fits to the EL spectra of the blue OLEDs. See text for more details.

multi-peak and the broadening of the spectrum are associated with the electronic transitions from the first excited state to the vibrational energy levels built on the ground state in the dopant molecule. As shown in Fig. 6, we identified the emission spectra to have five peaks and accordingly established energy levels of the dopant molecule, which will be discussed later as well.

It is obvious that the intensity of the second peaks is weakened as going from device A to D in spite of devices A and B showing a similar intensity. Accordingly, the relative intensity of the first peak area to the second area follows the order of devices D, C, B, and A. Interestingly, this feature is similar to the characteristics of spectral overlaps between the absorption of the dopant and the PL emission of the host molecules. This correlation, therefore, suggests that electronic excitation transitions with the second EL peak energy are easily affected by the dipole-dipole interactions by the host molecules.

Table I summarizes the results of multi-Gaussian fits for the two peaks that correspond to the strong electromagnetic wave transitions in the BCzVBi molecule. The transition energies of the two peaks were found to be 2.82 and 2.67 eV, respectively, and the relative intensities vary from 1:0.88 to 1:0.55 from devices A to D. It is clear that device D has the weakest intensity of the second peak among the devices. This phenomenon suggests that the low intensity of the second transition in device D should be attributed to the strong energy transfer between BCzVBi and TPBi molecules since this dopant-host combination has the largest spectral overlap as shown in Fig. 5. Further, we recognize that the first-second peak ratios follow the order of devices D, C, B, and A. This feature is similar to the spectral overlap order between the absorption of the dopant and the PL emission of the host molecules. As a consequence, we suggest that electronic excitation transition with the secondary EL peak energy, which corresponds to the second excitation mode of vibrations built on the ground state of the dopant molecule, has a strong correlation to the dipole-dipole interactions between the host and dopant molecules.

Before discussing how the relative ratios of the two transitions relate to the characteristics of the current-voltage and luminance-current in the devices, the presence of the shoulder spectrum peak at 530–720 nm in EL spectrum of device B shown in Fig. 7 should be explained.

To investigate the origin of this bump, it is essential that the pure EL spectrum and the shoulder spectrum should be separated. By noticing that the spectra of devices B and C are very similar in structure above 500 nm region, we applied the EL data of device C for the data of pure EL of

TABLE I. Transition energies and relative intensities of two major peaks in EL spectra. The numbers in parentheses denote the errors in the last digit.

| Device; host | $E_1$ (nm; eV)    | $E_2$ (nm; eV)     | $I_1: I_2$ |
|--------------|-------------------|--------------------|------------|
| A; NPB       | 440.3(4); 2.82(1) | 465.1(5); 2.67(1)  | 1:0.88     |
| B; DPVBi     | 441.3(3); 2.81(1) | 465.5(6); 2.67(1)  | 1:0.85     |
| C; MADN      | 440.4(8); 2.82(2) | 467.1(11); 2.66(2) | 1:0.69     |
| D; TPBi      | 441.2(6); 2.81(2) | 465.7(12); 2.67(2) | 1:0.55     |

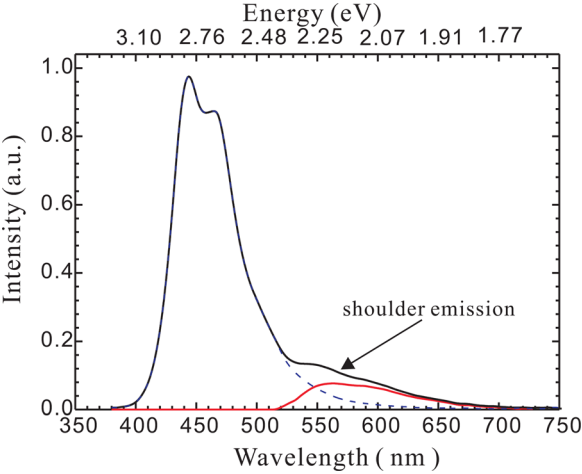


FIG. 7. (Color online) EL spectrum of BCzVBi doped in DPVBi (device B).

device B for this region. By this treatment we could isolate the shoulder spectrum from the EL of device B. Figure 7 represents the pure EL spectrum of device B with a dashed line and the isolated shoulder spectrum with a red curve. Our next step is to analyze the shoulder spectrum by multi-peak fits with a Gaussian function to identify the transition energies. We provide a fitting result for the shoulder spectrum along with the EL analysis in Fig. 8.

As shown in Fig. 8, the shoulder spectrum has a range with the energy of 2.3–1.8 eV and four peaks at 2.25, 2.11, 1.96, and 1.84 eV, respectively. The peaks imply that there are other electronic transitions in this host-dopant system. To gain physical insights into the origin of the transition energies, we investigated the energy levels of BCzVBi from the present EL data and then compared them to the LUMO-HOMO energy level of DPVBi. The energy levels derived from the present measurements and the corresponding electronic transitions for BCzVBi and DPVBi are shown in Fig. 9.

We now discuss the molecular level structure of the dopant BCzVBi and then the transitions associated with the

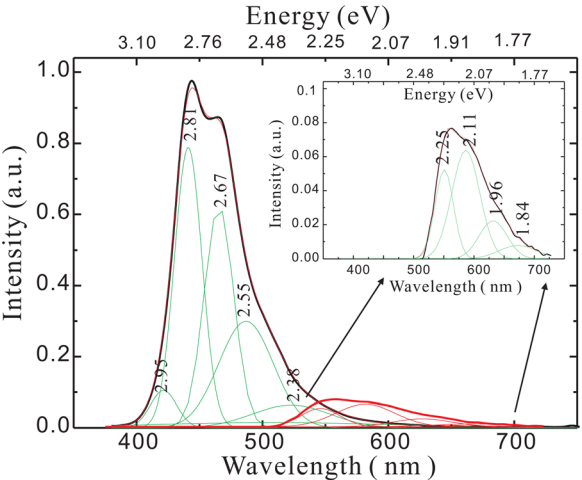


FIG. 8. (Color online) Multi-peak fits with Gaussian function for EL spectrum of BCzVBi in device B. Inset represents the shoulder region of EL spectrum and a corresponding Gaussian fit. It should be noted that the peaks are denoted as electron volts in energy scale.

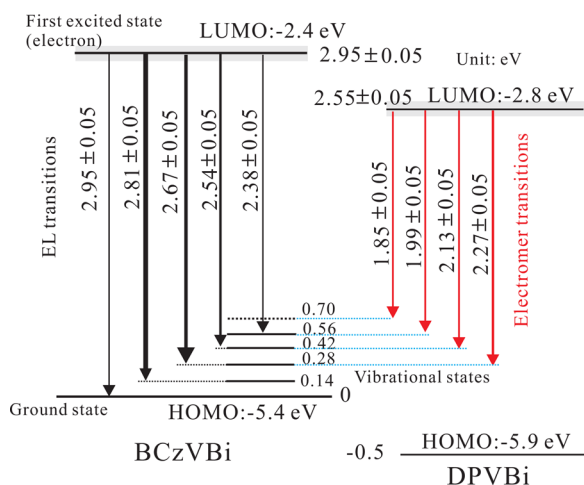


FIG. 9. (Color online) Energy level scheme of BCzVBi and DPVBi molecules as deduced from the present work. It should be noted that the energy levels for DPVBi represent relative to the ground state of BCzVBi. Energies are given in electron volts.

shoulder peaks. As shown in the energy level scheme of BCzVBi on the left side in Fig. 9, the EL peaks can be easily explained as resulting from the transitions from the first excited state  $S_1$ , namely the LUMO level, to the vibrational energy levels built on the ground state  $S_0$ . The energy of 0.14 eV between the peaks corresponds to the vibrational spacing energy,  $\Delta E_{vib} = h\nu$ , where  $h$  is the Planck constant and  $\nu$  is the phonon frequency. It should be emphasized that the vibrational states are not due to electron excitations but phonon excitations by molecular harmonic vibrations. The energy of such a harmonic oscillator is quantized,  $E_n = (n + 1/2)h\nu$ , where  $n$  is a quantum number  $n = 0, 1, 2, 3, \dots$ . The energy levels are equally spaced by an amount  $h\nu$ , the amount of energy absorbed or emitted by the oscillator when it is excited and relaxed to the neighboring energy levels. As already mentioned, we take the excited state  $S_1$  to be  $E_1 = 2.95 \pm 0.05$  eV, which corresponds to the bandgap for BCzVBi molecule, by assuming the uncertainty of the present EL transitions to be  $\pm 0.05$  eV. See Fig. 4. Here a weak transition with 2.95 eV might be associated with the transition from the LUMO  $S_1$  to HOMO  $S_0$  levels, namely  $S_1(n=0) \rightarrow S_0(n=0)$ .

As illustrated in Fig. 9, the relationship between the energy levels of BCzVBi and DPVBi explains that the shoulder emission should be associated with the intermolecular transitions from the first excited state of DPVBi to the vibrational energy levels built on the ground state of BCzVBi. In other words, this longer wavelength emission arises from this excited state complex, which is referred to as an electromer.<sup>8,9</sup> Generally, an electromer emission appears to be lower in energy and thus results in a redshift of the EL emission. Consequently, the shoulder emission at lower energies for device B is due to the formation of an electromer, which is a charge-transfer complex between the excited state of host DPVBi and dopant BCzVBi molecules. The emitted photon energy of the electromer is decreased by the amount of

$$\Delta = E_{LUMO}^{BCzVBi} - E_{LUMO}^{DPVBi} = 0.4 \text{ eV}$$

compared to that of pure EL transitions. The intensity of the electromer emission is found to be about 10% relative to that of the EL spectrum. The presence of such an electromer associated with intermolecular interactions has a significant effect on the quality of luminescence in the devices as we will discuss this point in the next section. Therefore, we conclude that device B with the DPVBi-BCzVBi system as an emissive layer should be avoided for high-efficiency OLEDs.

## B. Characteristics of light efficiency

Figure 10 shows the measured current density and luminance at the applied bias voltages for the devices. Generally, as the OLED current increases so does the injected carrier concentration, and thus the rate of recombination and hence the output light intensity. The increase in the output light power, however, is not linear with the current as shown in Fig. 10. Some important phenomena of the characteristics for the current and the output light luminance at the applied bias should be noted: Firstly, current efficiency is divided into two groups such as devices A and D with higher intensity than that of devices B and C; secondly, luminance of devices B and D is higher in intensity than that of devices A and C; lastly, the efficiency of current and luminance for device A reflects a striking contrary to each other.

As a consequence, device D has the highest current and luminance performance among the present devices. This result is consistent with the largest spectral overlap between the absorption and emission spectra for device D. It should

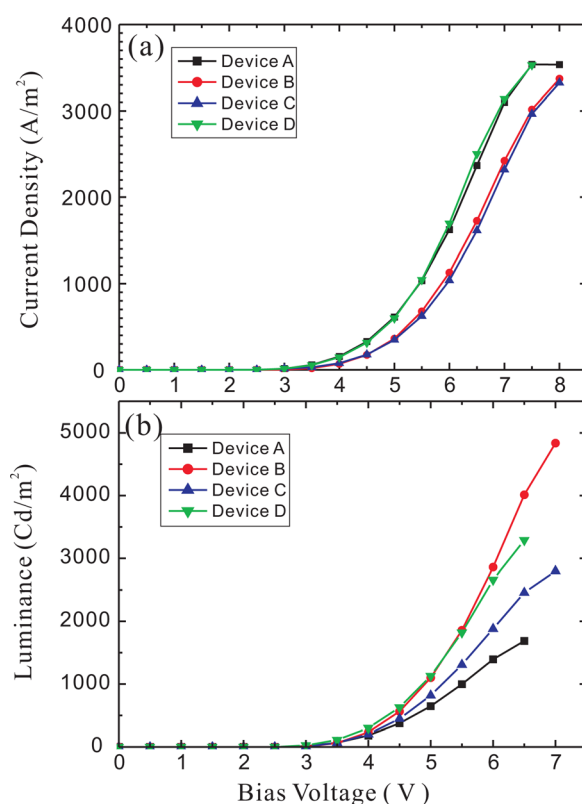


FIG. 10. (Color online) (a) Current density and (b) luminance as a function of bias voltage for the devices. Notice that all data are given in Systeme International units.

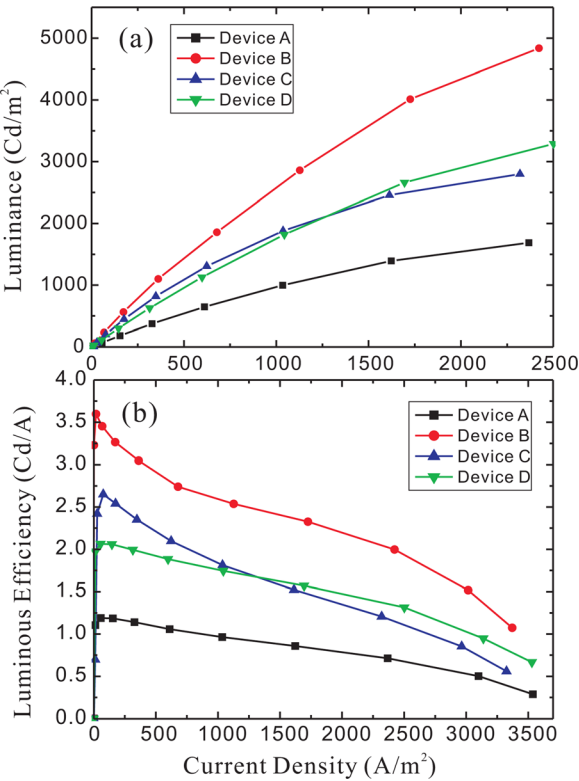


FIG. 11. (Color online) (a) Characteristics of luminance vs current density and (b) luminous efficiency vs current density for the devices.

be emphasized that device D has the weakest second transition intensity. We observe that the larger the spectral overlap for the present dopant-host systems, the weaker the second peak intensity. Interestingly, except for device B, luminance of devices D, C, and A is in direct proportion to the order of the spectral overlap size as well as the intensity ratio of the first and second peaks. This indicates that the second peak, namely, the transition to the  $n=2$  vibrational level can be readily affected by the molecular interactions between the dopant and host materials. It is evident that the molecule TPBi is the most promising host material leading to highly efficient EL emission for the devices. Although device B with the host of DPVBi shows the highest luminance performance, such an enhancement seems to be caused by the increase of emission due to the electromer.

Figure 11 shows plots of luminance and luminous efficiency as a function of current density for the devices. Again,

TABLE II. Characteristics of the devices for physical properties; EL spectrum, current density, luminance, and luminous efficiency.

| Physical properties                                     | Efficiency order of the devices              |
|---------------------------------------------------------|----------------------------------------------|
| Spectral overlap between the host and dopant            | $D > B \approx C > A (D > C > A)^a$          |
| Intensity ratios between the two peaks in EL; $I_1/I_2$ | $D > B \approx C > A (D > C > A)^a$          |
| Current density at applied bias                         | $A \approx D > B \geq C (A \approx D > C)^a$ |
| Luminance at applied bias                               | $B \approx D > C > A (D > C > A)^a$          |
| Luminous efficiency                                     | $B > C \approx D > A (C \approx D > A)^a$    |

<sup>a</sup>Excluding device B for neglecting effect of the emission due to the electromer.

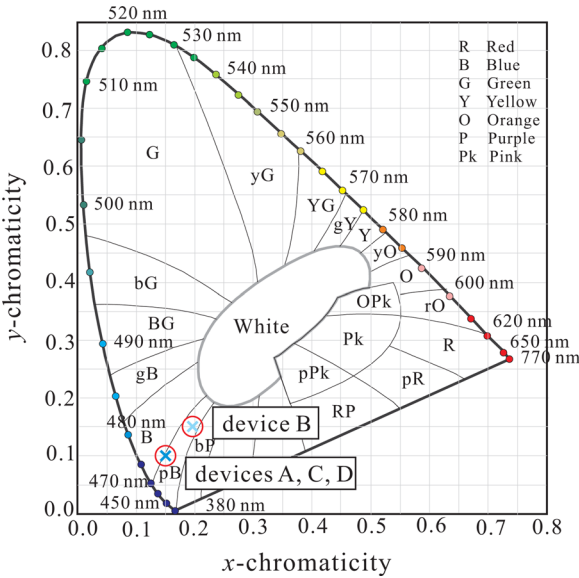


FIG. 12. (Color online) CIE chromaticity diagram along with the locations of blue emission from the present blue OLEDs.

we see that device B has the highest luminous efficiency in spite of the lower current density at applied bias voltages. Such a discrepancy between the current density and the emission efficiency for device B can be explained in terms of the formation of the electromer that drives to enhance output light emission. Namely, the longer wavelength emission due to the electromer significantly makes an impact on the luminance of the dopant. Furthermore, this impact leads to the poor blue coloration.

For summarizing, we present characteristics of the devices for the key features like spectral overlap, EL spectrum, current, and luminous efficiency in Table II. It is clear except for device B that the spectral overlap and second peak intensity have a large correlation with the luminous efficiency for the devices. As a consequence, device D has the best performance for the energy transfer and emission efficiency in the devices. The molecule of TPBi, therefore, is the best host material suitable for a high-efficient emission for the dopant of BCzVBi. The present result shows that the luminance of a dopant increases in direct proportion to the strength of spectral overlap responsible for electron energy transfer between the host and dopant molecules.

### C. Color chromaticity of the devices

Figure 12 depicts the color coordinates of CIE (Commission Internationale de l'Éclairage)<sup>23</sup> along with blue points obtained from the devices. Table III provides the CIE chromaticity coordinates including the electrical and optical properties for the devices. As shown in Fig. 12, the color coordinates of devices A, C, and D are located in purple blue point (0.15, 0.10) while device B is far off such a pure blue coordinate.

We can easily understand that such a color deviation of device B should be attributed to a red-shift emission due to the formation of an electromer. Again, device B indicates that the BCzVBi and DPVBi combination should be avoided as an emission layer for an OLED.

TABLE III. Electrical and optical properties of the blue OLED devices.

|                                                         | Device A (NPB) <sup>a</sup> | Device B (DPVBi) <sup>a</sup> | Device C (MADN) <sup>a</sup> | Device D (TPBi) <sup>a</sup> |
|---------------------------------------------------------|-----------------------------|-------------------------------|------------------------------|------------------------------|
| Current density (A/m <sup>2</sup> ) at 4.5 V            | 327                         | 172                           | 176                          | 314                          |
| Luminance (Cd/m <sup>2</sup> ) at 2000 A/m <sup>2</sup> | 1510                        | 4300                          | 2620                         | 2900                         |
| Luminous efficiency (Cd/A) at 4.5 V                     | 1.1                         | 3.3                           | 2.6                          | 2.0                          |
| CIE (x,y) coordinates at 4.5 V                          | (0.15, 0.11)                | (0.19, 0.15)                  | (0.15, 0.10)                 | (0.15, 0.10)                 |

<sup>a</sup>Host materials in the emissive layer.

IV. CONCLUDING REMARKS

To investigate optical properties of blue light emission in the host-dopant system, the organic light-emitting devices were fabricated with an activator of BCzVBi doped into the various host materials such as NPB, DPVBi, MADN, and TPBi. Through spectroscopic analysis based on multi-peak fits to the emission spectra, we found that the energy transfer intensity due to molecular interactions in the emissive layer has a profound effect on current density, luminance, luminous efficiency, and CIE color coordinates for the devices. More importantly, we established the energy level scheme of BCzVBi and identified the formation of an electromer in the BCzVBi and DPVBi system. This complex excimer formed by the dopant-host molecules significantly affects the optical and electrical properties for the device. We expect that the present quantitative analysis of the energy levels of BCzVBi makes a positive impact on device optimization for high-efficiency blue OLEDs.

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