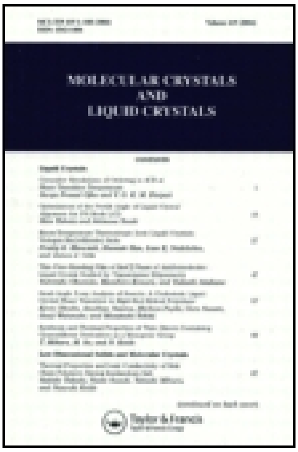

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Novel Optical and Electroluminescent Characteristics of Organic Multiple Quantum Well Structure Utilizing Fluorescent Dye Molecules

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NOVEL OPTICAL AND ELECTROLUMINESCENT CHARACTERISTICS OF ORGANIC MULTIPLE QUANTUM WELL STRUCTURE UTILIZING FLUORESCENT DYE MOLECULES

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Abstract Novel optical and electroluminescent characteristics of organic multiple-quantum-well (MQW) structure have been discussed comparing with MQW structure which consists of cyclopentadiene derivatives (PPCP) and aromatic diamine (TPD) and that of aluminum quinoline (Alq_3) and aromatic diamine (TPD). Energy transfer of excitons has been found in both MQW structures of PPCP/TPD and Alq_3/TPD systems. Unique electroluminescent characteristics with emission spectrum has been discussed in PPCP/TPD MQW system comparing with that of Alq_3/TPD MQW system.

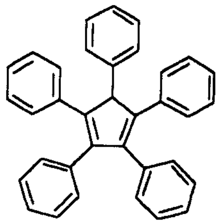
INTRODUCTION

Organic electroluminescent (EL) diodes[1-7] have attracted much attention because of their potentiality in materials and device processes. Tang *et al.*[1] developed a very efficient fluorescent material (tris(8-hydroxyquinoline) aluminum; Alq_3) and demonstrated low-voltage-driven EL diodes using Alq_3 as an emitting material. On the other hand, in inorganic III-V compound semiconductors MQW structures have demonstrated many unique optical and electrical characteristics compared with that in conventional bulk materials. So *et al.*[8,9] reported fabrication and optical characteristics of a crystalline organic MQW structure by using organic molecular beam deposition, and we reported anomalous optical characteristics of organic MQW structure of Alq_3/TPD[10-12] and other organic dye materials.[13-14]

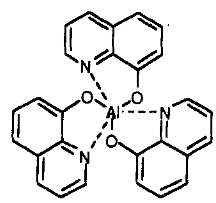
In this paper, we discuss novel optical and electroluminescent characteristics of organic MQW structure utilizing organic dye molecules, mainly on PPCP/TPD system comparing with that on Alq_3/TPD system.

EXPERIMENTAL

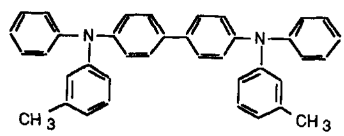
An organic superlattice structure was grown by organic molecular beam deposition onto two kinds of substrate. A quartz substrate was used for optical measurement, and an indium-tin-oxide (ITO) coated glass substrate for the EL diode. The base chamber pressure was kept under 10^{-4} Pa during deposition. The powders of Alq_3, cyclopentadiene derivative (1,2,3,4,5-pentameters-1,3-cyclopentadiene; PPCP) and aromatic diamine (N,N'-diphenyl-N,N'-(3-methyl phenyl)- 1,1'-biphenyl-4,4'-diamine; TPD) were loaded into separate Knudsen cells. The cells were subsequently heated up to sublimate and the growth rates were determined by an oscillating quartz thickness monitor. Molecular structures of organic materials used in this study are shown in Fig. 1.



(a)1,2,3,4,5-pentaphenyl-1,3-cyclopentadiene, (PPCP)



(b)tris(8-hydroxyquinoline) aluminum (Alq_3)



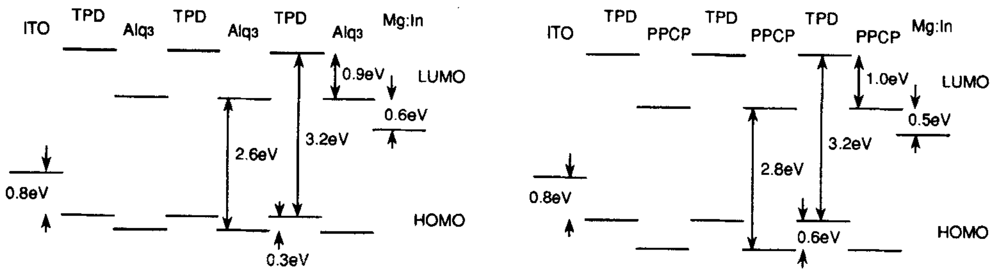
(c) N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD)

FIGURE 1 Molecular structures of organic materials used in this study.

A series of multilayer samples consist of alternate layers of Alq_3 or PPCP and TPD. The layer structure was determined by low angle X-ray (Cu-Kα line) diffraction.[10] Both MQW structures exhibited the X-ray diffraction signals from periodical layer structures. The EL diode consists of the superlattice structure sandwiched by an ITO-coated transparent electrode as the positive bias side and In-containing Mg (Mg:In) electrode as the negative bias side. The Alq_3 or PPCP layer provides contact with the Mg:In electrode and the TPD layer with the ITO electrode. The emission area is 2mm square.

OPTICAL CHARACTERISITICS OF ORGANIC MQW STRUCTURE

An energy band diagram for the MQW structure was estimated from optical absorption and cyclic voltammetry to determine the energy gaps and the HOMO states of the materials, respectively. Taking these results into consideration, it was found that LUMO state of PPCP are surrounded by the TPD barriers whereas the HOMO state of TPD by the PPCP barriers in PPCP/TPD system as shown in Fig. 2(a). Similarly, energy band diagram for Alq3/TPD system are estimated as shown in Fig. 2(b).



(a) PPCP/TPD system (b) Alq3/TPD system

FIGURE 2 Energy band diagrams of MQW structures of PPCP/TPD and Alq3/TPD systems.

The photoluminescence (PL) spectra with the excitation wavelength of 300nm are shown in Fig. 3 for the samples with MQW structure (TPD 5.5nm, PPCP 5.5nm, 20 periods; solid line) and heterostructure (TPD 55nm, PPCP 55nm, 1 period; dashed and broken lines). The emission intensity is normalized in this figure. Single layer of the deposited PPCP (100nm thickness) exhibits blue fluorescence at 457nm. The emission peak wavelength of the PL spectrum of the MQW structure was at 457nm and corresponded to that of PPCP, while the emission peak wavelength of the PL spectrum of the heterostructure was at 426nm. Since the single layer of TPD shows fluorescence at around 400nm, the PL spectrum of the heterostructure is the one overlapped with those of TPD and PPCP. Most of the excitons generated in the PPCP or TPD layers recombine in the own layers and result in the light emission. PL spectra of the heterostructure excited by 260nm light is shown in Fig.3 (dotted line). The PL spectrum shift to longer wavelength compared with that excited with 300nm. Photogenerated excitons are transferred to PPCP layers and result in the emission. In the heterostructure system, PL was observed from both the layers, and the emission changes with the excitation light.

On the other hand, in the MQW structure, emission from TPD is not observed and the spectrum does not change with the excitation wavelength. That is, only the emission from PPCP layer was observed from the MQW structure. From these results, it should be mentioned that the excitons are confined in the PPCP layer in the MQW structure, and the excitons generated in the TPD layer are transferred into the PPCP layer efficiently and result in the emission in the PPCP layer.

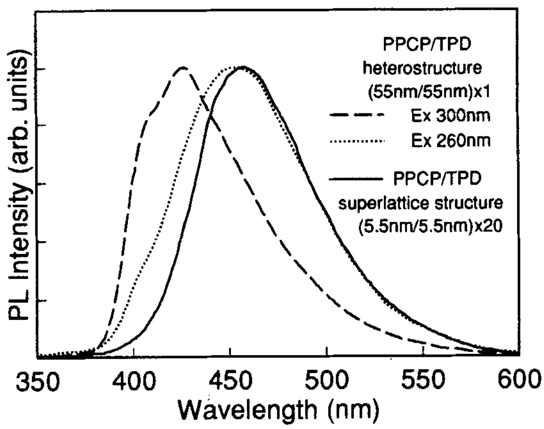


FIGURE 3 Photoluminescence spectra of PPCP/TPD MQW and heterostructure systems.

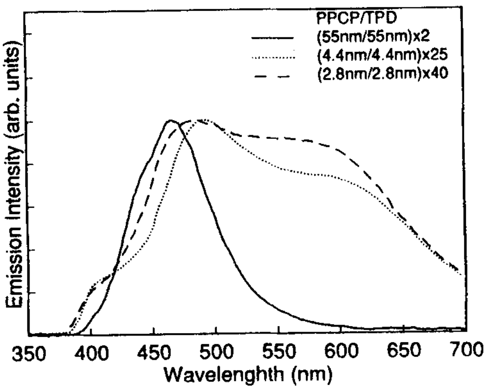
In the MQW structure of Alq3/TPD system, both the photoluminescence from TPD and Alq3 has been observed. However, emission from Alq3 increases as the layer thickness decreases. Similar energy transfer has been observed in the Alq3/TPD MQW system, where the strong emission has been observed from Alq3 in the short-period MQW system.

ELECTROLUMINESCENT CHARACTERISTICS OF ORGANIC MQW STRUCTURE

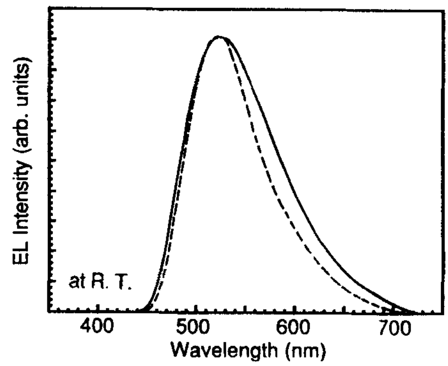
Current-voltage characteristics of the EL diodes with heterostructure and MQW structure show strong rectifying characteristics in both PPCP/TPD and Alq3/TPD systems. Since the energy band structures of the MQW structures are same in both cases, similar current-voltage characteristics should be obtained.

In Fig. 4, electroluminescence spectra for the diodes with MQW structure are shown. The emission spectrum of the MQW structure with long-period MQW (TPD 55nm, PPCP 55nm, 2 periods) corresponds to that of the diodes with heterostructure which shows the emission at around 460nm. However, the short-period MQW (TPD 4.4nm, PPCP 4.4nm, 25 periods; TPD 2.8nm, PPCP 2.8nm, 40 periods) show emission at around 600nm besides the emission at around 460nm. Since the emission at 600nm becomes stronger with decreasing the layer thickness of PPCP, it relates to the thin PPCP layer or the interface of PPCP and TPD.

In the short-period structure, the interface is interpreted to play larger role and the interaction between PPCP and TPD layers becomes larger against the emission from the bulk PPCP. In the case of the diodes with long-period structure, the injected carriers result in the emission at around 460nm. However, in the short-period structure, recombination through another energy levels located below LUMO or above HOMO states results in the emission at around 600nm. As we have observed no emission at 600nm at low temperature (77K), there should exist energy barrier to transit to the newly created energy levels. The new energy levels may be created by the interaction of the PPCP and TPD layers. The original bulk energy level is lost by the interaction of the energy levels between PPCP and TPD. The exciplex formation has to be also considered.



(a) PPCP/TPD system



(b) Alq3/TPD system

FIGURE 4 Electroluminescent spectrum of MQW systems.

On the other hand, in the case of Alq3/TPD MQW system, as shown in Fig. 4(b), the emission spectrum from MQW structure shift to the shorter wavelength and the full-width at half-maximum of the emission from MQW structure decreases as the layer thickness become thinner. In this MQW system, the emission spectrum is not broadened in the short-period MQW structure. The solid curve shows the EL spectrum from heterostructure with 53.6nm-thick Alq3 and 53.6nm-thick TPD, and the dashed curve that from MQW structure with 8 periods of 9.5nm-thick Alq3 and 9.5nm-thick TPD.

CONCLUSIONS

The experimental results are summarized as follows.

1) The photoluminescence spectrum with organic MQW structure of PPCP and TPD exhibits a peak emission from PPCP. Since the spectrum does not depend upon the excitation light, it is interpreted that the energy transfer from TPD to PPCP has been

conducted, and the excitons are confined in the PPCP layer. Similar energy transfer has been observed in Alq3/TPD MQW system, where the strong emission has been observed from Alq3 layer in the short-period MQW structure.

2) The electroluminescence spectra of the diodes with PPCP/TPD MQW structure exhibits only from PPCP. In the short-period structure, newly created emission at longer wavelength is observed besides the original emission of PPCP. This is caused by the interaction or the interface of thin PPCP and TPD layers. In the MQW structure the carriers or excitons are transferred to the PPCP layer resulting in the emission from PPCP layer. On the other hand, in the Alq3/TPD MQW system, spectral narrowing and emission energy shift to higher energy have been observed. In the MQW system of Alq3/TPD, interaction between Alq3 and TPD are considered to be very small and the emission from Alq3 has been kept even in the short-period MQW system.

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