Vol 17 No 8, August 2008
1674-1056/2008/17(08)/3097-06

Chinese Physics B

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White organic light-emitting device with both phosphorescent and fluorescent emissive layers

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(Received 29 October 2007; revised manuscript received 29 November 2007)

This paper reports the fabrication of novel white organic light-emitting device(WOLED) by using a high efficiency blue fluorescent dye *N*-(4-((E)-2-(6-((E)-4-(diphenylamino)styryl)naphthalen-2-yl)vinyl)phenyl)-*N*-phenylbenzenamine (*N*-BDAVBi) and a red phosphoresecent dye bis (1-(phenyl) isoquinoline) iridium (III) acetylanetonate ($Ir(piq)_2(acac)$). The configuration of the device was ITO/PVK:TPD/CBP: *N*-BDAVBi /CBP/ BALq: $Ir(piq)_2(acac)$/BCP/Alq_3/LiF:AL. By adjusting the proportion of the dopants (*N*-BDAVBi, $Ir(piq)_2(acac)$) in the light-emitting layer, white light with Commission Internationale de l'Eclairage (CIE) coordinates of (0.35, 0.35) and a maximum luminance of 25350cd/m^2 were obtained at an applied voltage of 22V. The WOLED exhibits maximum external quantum and current efficiency of 6.78% and 12cd/A respectively. By placing an undoped spacer CBP layer between the two light-emitting layers and using BCP as hole blocking layer, the colour stabilization slightly changed when the driving voltage increased from 6 to 22 V.

Keywords: WOLED, colour stability, *N*-BDAVBi
PACC: 7860F, 7280L

1. Introduction

White organic light-emitting devices (WOLEDs) are of interest due to their potential importance for the application, such as backlights in liquid crystal displays (LCDs), solid state lighting (SSL), and full-colour displays combined with colour filters.[1,2] In order to find widespread use, the WOLED should be efficient and possess good colour stability to applied voltage. Recently, prompted by the synthesis of red, green and blue phosphorescent dyes, more and more highly efficient phosphorescent small molecular and polymer-based WOLEDs have been reported.[3,4] White light emission is usually obtained from double-emissive-layer because of its less complexity in fabrication process. In general, OLED could exhibit white emission spectrum comprising tricolour or two complementary colours.[5] Whereas, white light emission which comes from the mixture of deep blue light and yellow light would result in the low efficiency. As a result, we employ a high efficiency greenish-blue fluorescent dye *N*-(4-((E)-2-(6-((E)-4-(diphenylamino)styryl)naphthalen-2-yl)vinyl)phenyl)-*N*-phenylbenzenamine (*N*-BDAVBi) and a red phosphorescent dye bis (1-(phenyl) isoquinoline) iridium (III) acetylanetonate ($Ir(piq)_2(acac)$. Through modulating their doping concentration and light emitting layer thickness, we can obtain a higher efficiency WOLED. To improve the colour stability, on the one hand an undoped spacer CBP layer was placed between the blue fluorophore and the red phosphor light-emitting layer, and on the other hand, BCP was sandwiched between the red emission and electron transporting layer.

2. Experimental

In this work, we report an efficient WOLED that has good chroma. The device consists of double light-emitting layers, in which 4,4′-*N*, *N*′-dicarbazole-biphenyl (CBP) doped with a fluorescent dopant *N*-(4-((E)-2-(6-((E)-4-(diphenylamino)styryl)naphthalen-2-yl)vinyl)phenyl)-*N*-phenylbenzenamine (*N*-BDAVBi) was used as blue emissive layer(EML), bis(2-methyl-8-quinolinolato)(para-phenylphenolato)aluminium(III) (BALq) doped with a phosphoresecent dopant of

bis(1-(phenyl)isoquinoline) iridium (III) acetylanetonate ($Ir(piq)_2(acac)$) was used as the red emissive layer(EML). A poly(*N*-vinylcarbazole) (PVK) doped *N*, *N*0-diphenyl-*N*, *N*0-bis (3-methylphenyl)-1, 1-biphenyl-4,40-diamine (TPD) layer was used as a hole transporting layer (HTL) while tris(8-hydroxyquinoline) aluminium (Alq_3) was used as an electron-transporting layer (ETL), and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) was used as exciton-blocking layer.

WOLEDs were fabricated on precleaned ITO glass substrates with a surface sheet resistance of about 30Ω/□. The PVK:TPD as the HTL was spin-coated onto the ITO glass substrate whose thickness was 80 nm, using chloroform as the solvent. The other

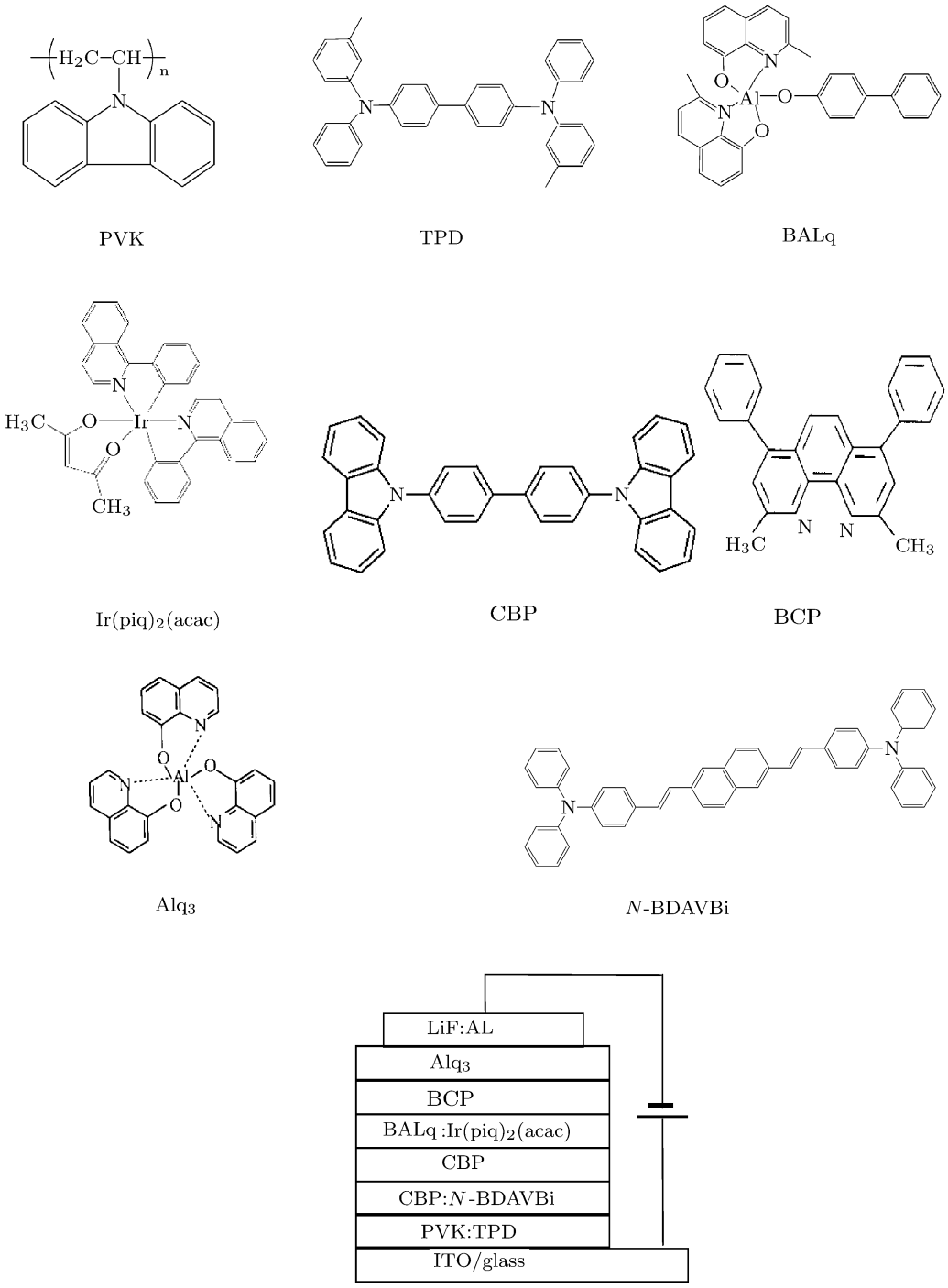


Fig.1. Molecular structures of the organic materials used and the configuration of the WOLED.

layers were successively deposited in high vacuum ($< 2\times10^{-4}$ pa) using a BOC Edwards Auto 500 Thermal Evaporation Coating System in the following sequence: CBP: *N*-BDAVBi (30 nm), CBP(5 nm), BAlq: Ir(piq)$_2$(acac) (15 nm), BCP(15 nm), Alq$_3$ (30 nm), LiF(8 nm), Al(1000 nm). The evaporation rates measured by a quartz crystal oscillator were 0.1–0.2 nm/s. The doping concentration of EML was controlled by monitoring the evaporation rates of each material. The emitting area was $3\times3\,\mathrm{mm}^2$. Figure 1 shows the construction of the WOLED with ITO/PVK:TPD/CBP: *N*-BDAVBi /CBP/ BALq: Ir(piq)$_2$(acac)/BCP/Alq$_3$/LiF:Al. and the molecular structures of the organic materials used in this work.

The electroluminescence (EL) spectra, brightness and CIE coordinates were recorded with a SpectraScan PR-650 (Photo Research). The quantum efficiency versus current density characteristics of the devices were measured using a purpose-built OLEDs testing system, comprising of a 2400 Sourcemeter (Keithley) and a 485 Picoammeter (Keithley).[6] All the measurements were carried out on unencapsulated devices at room temperature in air.

3. Results and discussion

It is universally acknowledged that the efficiency and colour stability of the WOLED could be improved by several approaches, such as the balance injecting of hole-electron, optimizing of layer thickness, appropriate doping concentration of dyes and so on. PVK is widely used as a HTL because of its excellent film-forming ability and high thermal stability. It is evident from Fig.2 that PVK has a lower HOMO (highest occupied molecular orbital) than that of TPD. Hence, the injection of holes is easier from ITO to TPD layer than to PVK under the same driving voltage. In our experiments, PVK:TPD was used as the hole-transporter layer, providing a compromise between the properties of the individual layers. With the electron-transport character, BALq, BCP and Alq$_3$ could make the injection and transport of electrons more easily. Furthermore, with the ambipolar transporter layer (CBP), carriers could be well balanced in each EML.

It was found that the changing of *N*-BDAVBi concentration from 1.0 wt% to 4.0 wt% had only a small

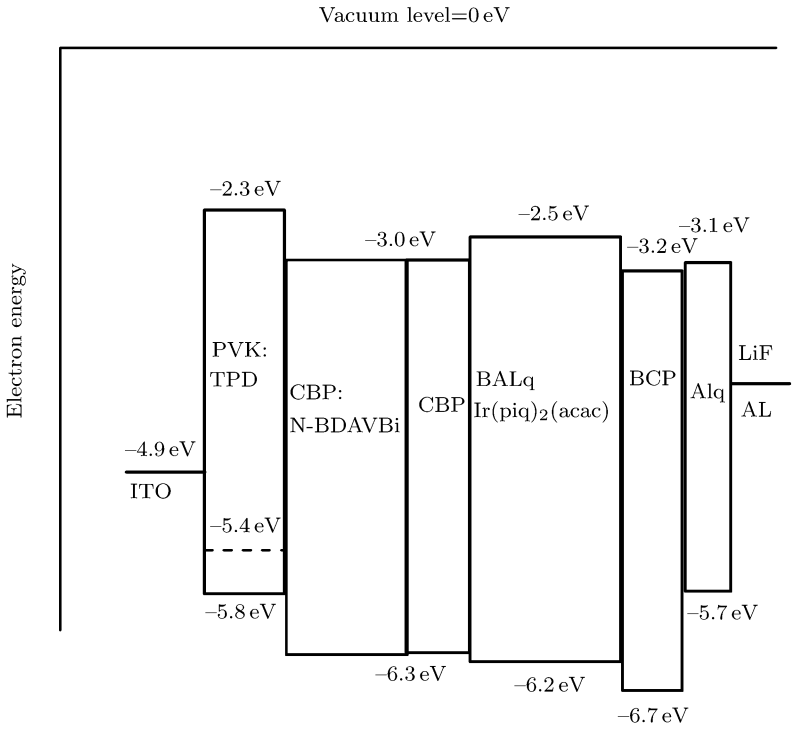


Fig.2. Energy level diagram of the used materials.

effect on the EL performance of the devices once the weight concentration of $Ir(piq)_2(acac)$ was above a certain value. This may be that *N*-BDAVBi undergoes significantly less molecular aggregation. Therefore, the EL performance of the *N*-BDAVBi-doped devices becomes insensitive to dopant concentration up to 4.0 wt%. As a consequence, it was expected that the white light emission could be obtained by adjusting the $Ir(piq)_2(acac)$ concentration.

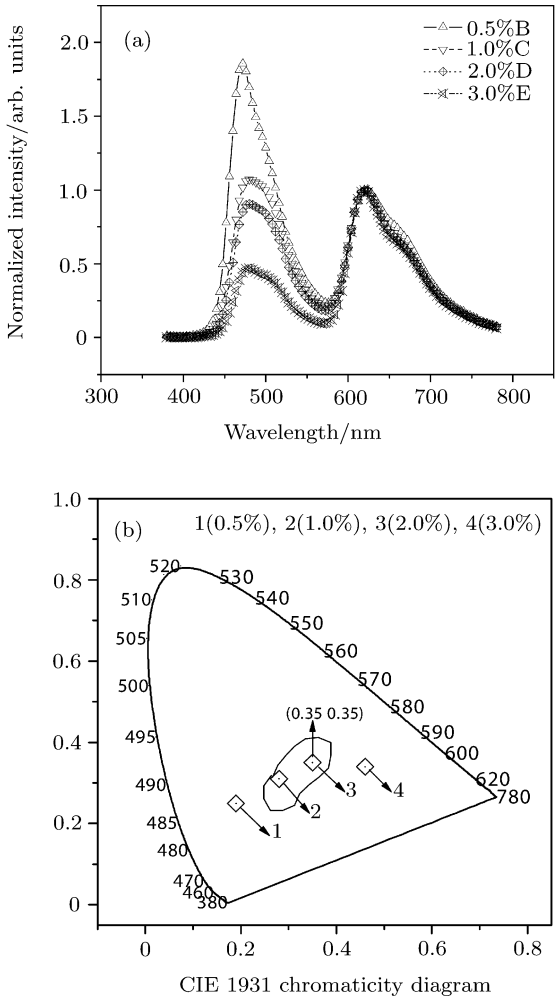


Fig.3. (a) Electroluminescence spectra for devices with different doping concentration of $Ir(piq)_2(acac)$. (b) The CIE coordinates for the devices of different doping concentration of $Ir(piq)_2(acac)$.

Figure 3(a) shows the EL spectra for devices with a fixed concentration of *N*-BDAVBi (3.0 wt%) and with variable concentrations of $Ir(piq)_2(acac)$(0.5%, 1.0%, 2.0% and 3.0%) at an applied voltage of 15 V, Fig.3(b) shows the corresponding CIE coordinates. White light emission was obtained from the device with *N*-BDAVBi and $Ir(piq)_2(acac)$ doping concentrations of about 3.0wt% and 2.0wt%, respectively. The figures also reveal that, the intensity of the *N*-BDAVBi emission peaks would decrease significantly once the concentration of $Ir(piq)_2(acac)$ is greater than 2.0wt%,. There are three possible explanations for this phenomenon: one reason is that CBP triplets with their characteristically long diffusion lengths (> 100 nm) may migrate into the spatially separated phosphor-doped region.[7,8] Second may be that the saturation of blue emission sites,[9] consequently the *N*-BDAVBi emission peaks slightly change. Last is that there is a little overlap between the emission spectrum of *N*-BDAVBi and the absorption spectrum of $Ir(piq)_2(acac)$ (Fig.4), so the energy can be transported by the Förster mechanism. In the Förster mechanism, dipole–dipole coupling results in a non-radiative transfer of the singlet excited-state energy from a donor molecule to an acceptor molecule. The overlap between the donor emission spectrum and the acceptor emission spectrum is an important criterion for Förster energy transfer to take place.[10]

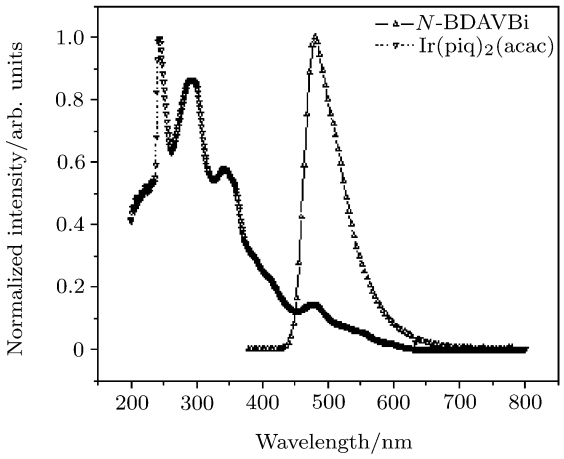


Fig.4. The emission spectrum of *N*-BDAVBi and the absorption spectrum of $Ir(piq)_2(acac)$.

Normalized EL spectra of the device with *N*-BDAVBi and $Ir(piq)_2(acac)$ doping concentrations of about 3.0wt% and 2.0wt% respectively, at various driving voltage are shown in Fig.5, which reveals the EL spectra and CIE coordinates of the device. It is evident that the EL spectra varies only slightly as the applied voltage changes from 6 V to 22 V. The corresponding CIE coordinates are (0.352, 0.349) at 6 V and (0.350, 0.345) at 22 V, which were much less than those observed for human. Generally, colour tuning happens in multilayer OLED due to the movement of the recombination zone for electrons and holes with applied voltage. The phenomenon has been theoretically studied by Chen and Meng.[11] According to their

theory, the recombination first concentrated in the red EML next to the BALq/BCP interface due to the relatively low electron mobility at low voltages, then it extends to the blue EML because the electric field strongly enhances the electron mobility as the voltage increases. However, the emission colour of this device is fairly stable with different driving voltage. One reason is that placing an undoped host spacer with a thickness larger than the Förster radius (> 3 nm) between the blue fluorophore and the phosphor prevents direct energy transfer from the blue dopant to the red phosphor.[12] Another is that the BCP layer is an effective hole blocking layer because of its relatively low HOMO energy (–6.7 eV in Fig.2). This layer will result in a greater accumulation of holes on the BALq side of the BALq /BCP heterojunction, providing an increased red emission. The device shows that high colour stability to applied voltage is the result of the combination of these processes.

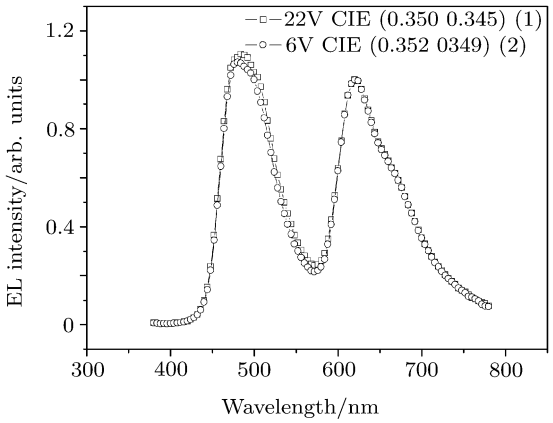


Fig.5. Normalized EL spectra of the OLED device (*N*-BDAVBi 3.0%, $Ir(piq)_2(acac)$ 2.0%) under various driving voltages.

Furthermore, this white OLED has a turn-on voltage of about 6 V, a maximum luminance of 25350 cd/m^2 at 22 V and a maximum external quantum 6.78% at 8.6 V, which are shown in Fig.6(a) and Fig.6(b).

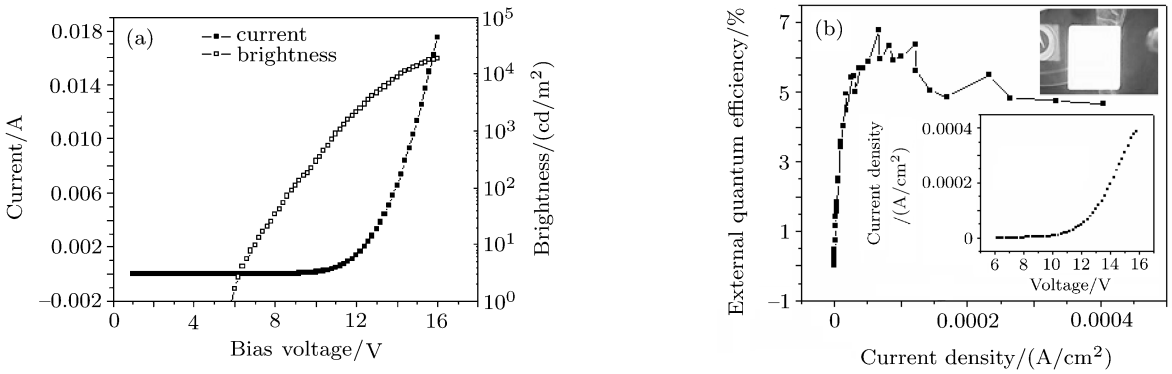


Fig.6. (a) The current-voltage-brightness curves of the device. (b) The external quantum efficiency–current density curves of the device.Inset:the driving voltage-current density curves and a photograph of the working device (3 cm×3 cm).

4. Summary

In summary, we demonstrate a WOLED based on a CBP host doped with a blue dye *N*-BDAVBi and a BALq host doped with a red dye $Ir(piq)_2(acac)$. Chromaticity of this device can be tuned by changing the concentration of dopants from their optimal values. To achieve a white emission in this device, the doping concentrations of *N*-BDAVBi and $Ir(piq)_2(acac)$ are optimized to 3.0wt% and 2.0wt%, respectively. The white device exhibits a good colour stability at different driving voltages (6–22 V). This OLED presents a maximum luminance of 25350cd/m^{2}with CIE coordinates of (0.35, 0.35) at 22 V and a maximum external quantum efficiency of 6.78% at 8.6 V. With the good colour stability in brightness work condition and simple device structure, the WOLED provides a route for the application in solid state lighting.

References

[1] Du P, Zhang X Q, Sun X B, Yao Z G and Wang Y S 2006 *Chin. Phys.* **15** 1370

[2] Zhang G H, Hua Y L, Wu K W, Wu X M, Yin S G, Hui J L, An H P, Zhu F J and Niu X 2007 *Acta Phys. Sin.* **56** 3559 (in Chinese)

[3] Xu Y H, Peng J B, Jiang J X, Xu W, Yang W and Cao Y 2005 *Appl. Phys. Lett.* **87** 193502

[4] Qin D S and Tao Y 2005 *Appl. Phys. Lett.* **86** 113507

[5] Wei F X, Cao J, Zhang X B, Liu X, Jiang X Y, Zhang Z L, Zhu W Q and Xu S H 2006 *Acta Phys. Sin.* **55** 2008 (in Chinese)

[6] Wu X M, Hua Y L, Luo J G, Feng X L and Li J 2002 *Chin. J. Lumin.* **23** 595 (in Chinese)

[7] Baldo M A and Forrest S R 2000 *Phys. Rev.* B **62** 10967

[8] Giebink N C, Sun Y and Forrest S R 2006 *Org. Electron.* **7** 375

[9] Li G and Shinar J 2003 *Appl. Phys. Lett.* **83** 5359

[10] Shaheen S E, Kippelen B, Peyghambarian N, Wang J F, Anderson D J, Mash E A, Lee P A, Armstrong N R and Kawabe 1999 *J. Appl. Phys.* **85** 7939

[11] Chen C H and Meng H F 2005 *Appl. Phys. Lett.* **86** 201102

[12] Sun Y R, Giebink N C, Kanno H, Ma B W, Thompson M E and Forrest S R 2006 *Nature* **440** 908