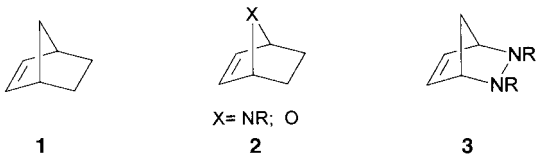
$\delta = 8.09$ (d, 6H, H4), 8.02 (s, 6H, H2), 7.91 (d, 6H, H7), 7.54 (m, 6H, H6), 7.46 (m, 6H, H5), 7.36(s, 6H, $C_6H_6$), 7.13–7.28 (m, 5H, $C_6H_5CH_3$), 5.70 (s, 12H, H8), 2.34(s, 18H; Me), 2.29 ppm (s, 3H; $C_6H_5CH_3$).

[20] Crystal data for $[Ag_3(MsTBim)_2](CF_3SO_3)\cdot(BPh_4)_2$: $M_r = 2132.35$, monoclinic, space group $C2/c$, $a = 25.866(5)$, $b = 18.041(4)$, $c = 22.125(4)$ Å, $\beta = 96.90(3)°$, $V = 10250(4)$ Å$^3$, $T = 293$ K, $Z = 4$, 8663 unique reflections measured, final $R1 = 0.0744$ and $wR2 = 0.1981$ for 5554 observed $[I > 2\sigma(I)]$ reflections. CCDC-183553 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge via www.ccdc.cam.ac.uk/conts/retrieving.html (or from the Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB2 1EZ, UK; fax: (+44) 1223-336-033; or deposit @ccdc.cam.ac.uk). The $CF_3SO_3^-$ ion is disordered along the $C_2$ axis.

## Two-Step, Stereoselective Hydrazidoarylation of 1,3-Cyclopentadiene**

Min-Liang Yao, Gunadi Adiwidjaja, and Dieter E. Kaufmann*

The first palladium-catalyzed reductive phenylation of norbornene (**1**) was published by Larock and Johnson[1] in 1989; since then the high synthetic potential of the hydroarylation[2] and especially its asymmetric variant[3] with bicyclic alkenes for the one-step construction of three asymmetric centers has induced a line of follow-up papers. This situation is especially true for the hydroarylation of the 7-aza- and oxabicyclic alkenes **2** (Scheme 1)[2b–d,3c] as this synthetic route even in the case of the asymmetric pathway leads directly to the biologically highly active alkaloid Epibatidine[4] and its analogues.




**1** **2** **3**

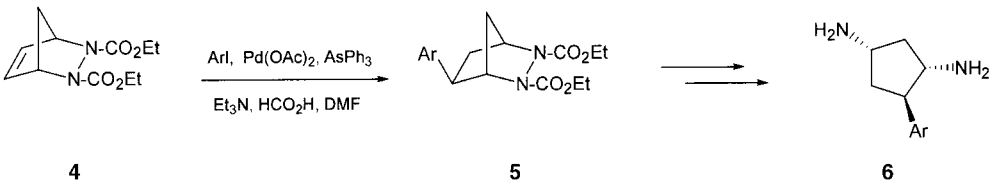
Scheme 1. (Hetero)norbornenes **1**–**3** as starting materials for hydroarylation reactions.

As we are interested in both the hydroarylation of bicyclic alkenes and, herein, in the use of these products in the stereoselective synthesis of substituted cyclopentane derivatives, we have investigated the hydroarylation followed by reductive cleavage of the easily accessible[5] 2,3-diazabicyclo[2.2.1]heptenes **3**,[6] in which the N–N[7] or C–N bond[8] represents an internal point of fracture.

Recently, we have reported the first palladium-catalyzed hydroarylation of the N,N′-diethoxycarbonyl-substituted derivative **4** of **3** with triethylamine as a base.[6] The following selective N–N bond cleavage opens a highly stereoselective way to the *trans*-4-aryl-*cis*-1,3-diaminocyclopentanes **6** (Scheme 2).

Encouraged by this initial result, we turned to the hydroarylation of the sterically more hindered and more rigid, tri- or tetracyclic Diels–Alder adducts of 1,3-cyclopentadiene with the very reactive azodienophiles 4-phenyl-1,2,4-triazoline-3,5-dione (**7a**) and 2,3-phthalazine-1,4-dione (**7b**). The reaction of **7a** with iodobenzene was chosen as a model system with which to optimize the reaction conditions; selected experiments are shown in Table 1.

We started with the reaction conditions ($Et_3N$, DMF, 65 °C, entry 1) which were optimal in case of the hydroarylation of **4**; besides 21% of the expected hydroarylation product **8a**, compound **9a** was formed in 9% as the product of a C–N cleavage reaction. Formally, the formation of **9a** is the result of a 1,2-hydrazidoarylation on the primarily employed 1,3-cyclopentadiene. From the $^1H$ and $^{13}C$ NMR spectroscopic data it appeared that the *trans* isomer **9a** was formed exclusively. The structural assignment is difficult in case of five-membered ring systems; however, the stereochemistry was unambiguously supported by an X-ray analysis (Figure 1).[9,10]




**4** **5** **6**

Scheme 2. Stereoselective synthesis of threefold substituted cyclopentane derivatives.

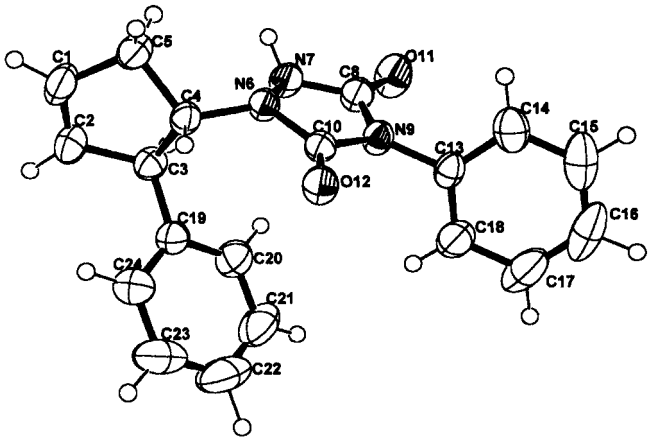



Figure 1. ORTEP plot of **9a**.

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[**] Palladium-catalyzed reactions, part 4; part 3 ref [6]. We thank BAYER AG, Leverkusen and Fond der Chemischen Industrie (Germany) for financial support.

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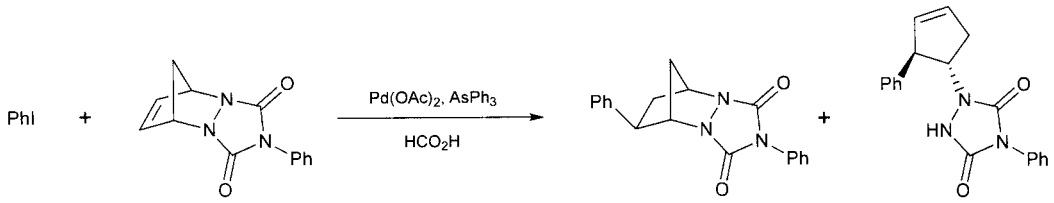
Table 1. Influence of base and temperature on the reaction of **7a** with PhI.



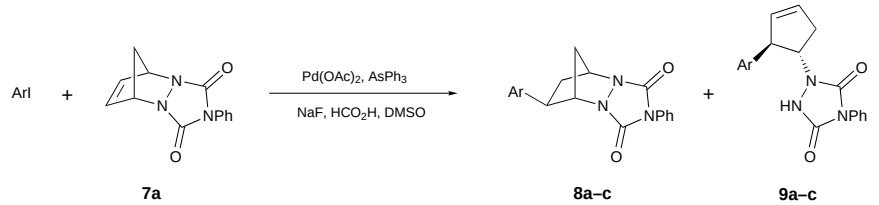

**7a** **8a** **9a**

| Entry | Reaction conditions[a] | Yield [%][b] **8a** | Yield [%][b] **9a** |
|---|---|---|---|
| 1 | $Et_3N$; DMF; 65 °C | 21 | 9 |
| 2 | $Et_3N$; DMF; 50 °C | 22 | 14 |
| 3 | $Et_3N$; DMF; 20 °C | 31 | 20 |
| 4 | NaOAc; DMSO; 20 °C | 31 | 28 |
| 5 | NaF; DMSO; 65 °C | 18 | 46 |
| 6 | LiCl; DMSO; 65 °C | 3 | 25 |

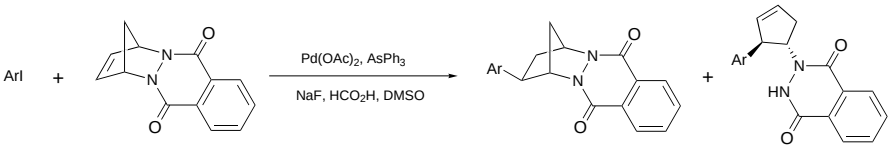
[a] Reaction conditions: 2.5 mol % $Pd(OAc)_2$, 11 mol % $AsPh_3$, alkene **7a** (1.0 equiv), PhI (1 equiv), base (3.5 equiv), $HCO_2H$ (3.0 equiv) in solvent (4 mL). [b] Yields of isolated product relative to alkene **7a**.

The low yield of **8a** is because of the low stability of the starting material **7a** towards the basic triethylamine at 65 °C; the cleavage by KOH[11] and hydrazine[12] is known. Lowering the reaction temperature from over 50 (entry 2) to 20 °C (entry 3) led to an overall yield of more than 50 %. On the one hand, the reactivity of **7a** in comparison with **4** is clearly remarkable, allowing an addition even at room temperature, however, on the other hand there is a distinct increase in the amount of by-product **9a** produced on decreasing the reaction temperature. However, we did not succeeded in raising the yield by using either a higher catalyst:ligand ratio or an excess of iodobenzene. On the other hand the use of inorganic bases proved to be successful: a heterogeneous mixture of sodium acetate and DMSO (entry 4) led to an approximately equidistribution of **8a** and **9a**. In recent years the application of alkali-metal fluorides has proved successful in palladium-catalyzed reactions.[13] With sodium fluoride in DMSO the conversion still proceeded very sluggishly at room temperature, while at 65 °C (entry 5), under acidic conditions (formic acid),[14] the hydroarylation was suppressed and **9a** was formed as the main product with an overall yield of 64 %. The use of lithium chloride as a base led almost exclusively to the formation of the formal hydrazidoarylation product **9a**; however the yield was dramatically reduced.

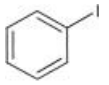
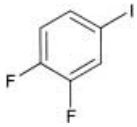
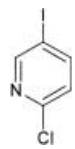
Table 2. Palladium-catalyzed reaction of (hetero)aryliodides with **7a/b**.




**7a** **8a–c** **9a–c**




**7b** **8d–f** **9d–f**

| Entry[a] | Alkene | ArI | Yield [%][b] **8a–f** | Yield [%][b] **9a–f** |
|---|---|---|---|---|
| 1 | **7a** |  | 18 (21) **8a** | 46 (9) **9a** |
| 2 | **7b** | | 3 **8d** | 67 **9d** |
| 3 | **7a** |  | 5 (18) **8b** | 63 (5) **9b** |
| 4 | **7b** | | 4 **8e** | 64 **9e** |
| 5 | **7a** |  | 7 (11) **8c** | 63 (7) **9c** |
| 6 | **7b** | | 6 **8f** | 69 **9f** |

[a] Reaction conditions: 65 °C, 2.5 mol % $Pd(OAc)_2$, $AsPh_3$ (11 mol %), alkene (1.0 equiv), ArI (1.5 equiv), NaF (3.5 equiv), $HCO_2H$ (3.0 equiv) in DMSO (4 mL). [b] Yields of isolated product, relative to alkene; the yields in parentheses were obtained under the reaction conditions in entry 1, Table 1.

After we had optimized the reaction conditions (Table 1, entry 5), we examined the application scope of the reaction for the polycyliclic diazaalkenes **7a/b** with three aryliodides (Table 2). Apparently, the use of sodium fluoride as a base

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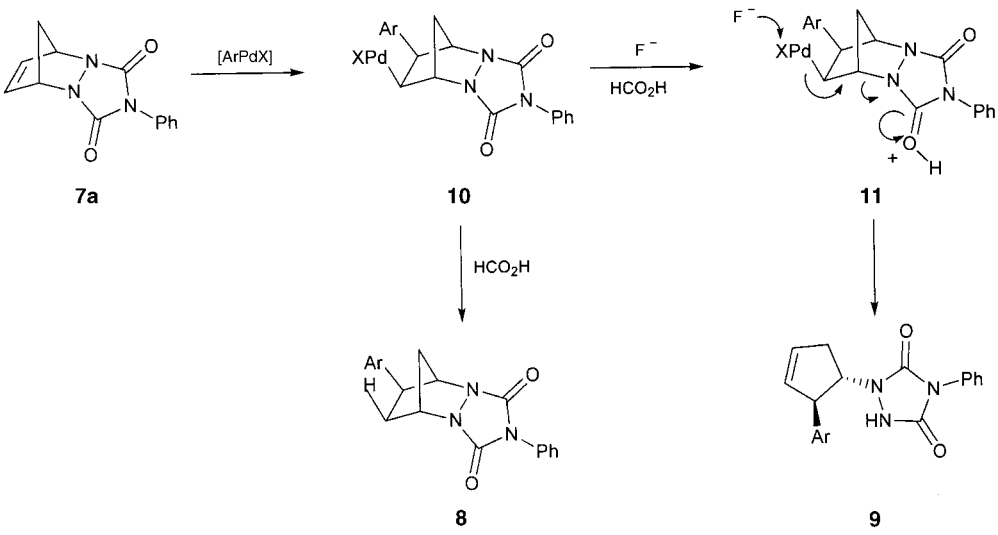
instead of triethylamine allows the selective formation of the opening products **9a–f** in good yields.

Several reaction paths are mechanistically conceivable. The cleavage of a C–N bond in diazabicyclic compounds through acid catalysis[15a] or heating[15b] is known. In a two-step process from **7a**, primarily a N-cyclopentadienyl-substituted phenylurazole could be formed, the subsequent, regioselective hydroarylation of which could lead to the *trans*-configured **9a**. The reaction of **7a** with formic acid or hydrochloric acid in DMF or DMSO at 65 °C, however, did not lead to the formation of even trace amounts of an opening product. Based on this result we suggest the mechanism outlined in Scheme 3.




Scheme 3. Mechanism of the palladium-catalyzed C–N cleavage of **7a**.

At the beginning, a *syn*-addition of a [ArPdX] species to the bicyclic C=C bond of **7a** occurs to give **10**. This intermediate can then be reduced to the hydroarylation product **8** either by formiates or even formic acid itself. Apparantly, electron-poor aromatic groups stabilize the intermediate palladium complex **10** towards reduction as effectively as the halide ions $F^-$ and $Cl^-$, so that a—preferentially acid catalyzed—competitive isomerization reaction can occur (see **11**) forming **9**. This methodology opens a new, simple access to *trans*-3,4-disubstituted cyclopentene derivatives, a class of intermediates and biologically active compounds that is not easily accessible by other pathways.

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[9] Crystal structure analysis: Structure solution with direct methods and full-matrix least-squares refinement procedure (SHELX97[10]). **9a** ($C_{19}H_{17}N_3O_2$, $M_r = 319.13$): monoclinic, space group $P2(1)/c$, $a = 17.3890(10)$, $b = 11.3510(10)$, $c = 19.9100(10)$ Å, $\beta = 156.100(10)$, $V = 1592.16(19)$ Å$^3$, $Z = 4$, $\rho_{calcd} = 1.332$ Mg m$^{-3}$, $T = 293$ K, 21 904 collected, 3656 crystallographic independent and 3473 reflexes mit $I > 2\sigma(I)$, $Mo_{K\alpha}$ radiation, $\lambda = 0.71073$ Å, $\theta_{max} = 27.50°$, $R[I > 2\sigma(I)]$: $R_1 = 0.0424$, $wR_2 = 0.1400$, $wR_2 = 0.1959$ (all data), 286 parameters, anisotropic thermal parameters, H-atoms isotropic. CCDC-173237 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge via www.ccdc.cam.ac.uk/conts/retrieving.html (or from the Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB2 1EZ, UK; fax: (+44) 1223-336-033; or deposit@ccdc.cam.ac.uk).

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[14] Typical experimental procedure: A solution of $Pd(OAc)_2$ (5.6 mg, 0.025 mmol) and $AsPh_3$ (33.7 mg, 0.11 mmol) in anhydrous DMSO (4 mL) was stirred under nitrogen at 65 °C for 15 min. Afterwards, the alkene **7a** (241 mg, 1.0 mmol), NaF (147 mg, 3.5 mmol), iodobenzene (306 mg, 1.5 mmol), and $HCO_2H$ (138 mg, 3.0 mmol) were added. The reaction mixture was stirred for another 16 h. After cooling to RT brine (50 mL) was added, the reaction mixture was extracted with ethyl acetate and dried with $MgSO_4$. The solvent was removed in

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vacuo and the residue was purified by column chromatography (silica, petroleum ether:ethyl acetate 9:1): mixture of **8a** (18%) and **9a** (46%). **8a**: colorless crystals, mp.: 174–175°C; $^1$H NMR (400 MHz, $CDCl_3$, TMS): $\delta = 1.92$–$2.21$ (m, 3H), 2.42–2.55 (m, 1H), 3.52–3.59 (m, 1H, HC-Ph), 4.72 (s, 1H, $H_{bridgehead}$), 4.80 (s, 1H, $H_{bridgehead}$), 7.16–7.55 ppm (m, 10H, $H_{aryl}$); $^{13}$C NMR (100 MHz, $CDCl_3$, TMS; Multiplicities of $^{13}$C NMR signals were determined by the DEPT sequence and are reported as (+) for CH or $CH_3$, (−) $CH_2$, and (o) for C): $\delta =$ 35.00 (−), 36.25 (−), 45.29 (+), 60.33 (+), 64.87 (+), 125.38 (+), 126.95 (+), 127.07 (+), 128.31 (+), 128.85 (+), 129.15 (+), 131.52 (o), 140.26 (o), 156.71 (o, C=O), 156.96 ppm (o, C=O); MS (EI): *m*/*z* (%) 320 (15, $[M^+ + 1]$), 319 (59, $[M^+]$), 214 (100), 143 (20), 142 (19), 115 (17), 104 (43), 91 (30); IR: $\tilde{\nu}_{max} = 3056$, 3011, 1776, 1716, 1598, 1501, 1415, 735, 697 cm$^{-1}$. HRMS: calcd for $C_{19}H_{17}N_3O_2$ 319.1321; found 319.1321. **9a**: Colorless crystals, mp.: 187–189°C. $^1$H NMR (400 MHz, $CDCl_3$, TMS): $\delta = 2.49$–$2.63$ (m, 1H), 2.77–2.92 (m, 1H), 4.03–4.07 (m, 1H, HC-Ph), 4.74–4.84 (m, 1H, HC-N), 5.73–5.83 (m, 1H, HC=), 5.90–5.96 (m, 1H, HC=), 7.17–7.49 (m, 10H, $H_{aryl}$), 9.07 ppm (br s, 1H, HN); $^{13}$C NMR ($CDCl_3$): $\delta = 35.26$ (−), 54.28 (+), 64.30 (+), 125.44 (+), 127.15 (+), 127.36 (+), 128.23 (+), 128.74 (+), 129.09 (+), 129.63 (+), 131.13 (o), 132.73 (+), 141.48 (o), 152.21 (o, C=O), 154.09 ppm (o, C=O); MS (EI): *m*/*z* (%) 320 (16, $[M^+ + 1]$), 319 (68, $[M^+]$), 215 (16), 214 (100), 143 (22), 142 (21), 104 (57), 91 (26); IR: $\tilde{\nu}_{max} = 3160$, 3059, 1772, 1687, 1595, 1493, 1378, 700, 676 cm$^{-1}$; HRMS: calcd for $C_{19}H_{17}N_3O_2$ 319.1321; found 319.1321.

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# Biomimetic Model of a Plant Photosystem Consisting of a Recombinant Light-Harvesting Complex and a Terrylene Dye**

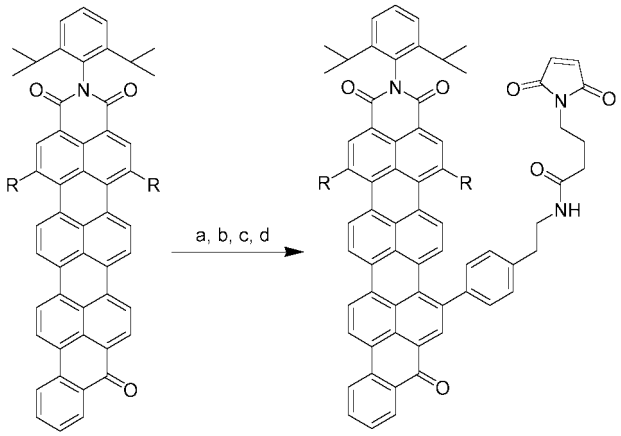
Henriette Wolf-Klein, Christopher Kohl, Klaus Müllen, and Harald Paulsen*

The light-harvesting chlorophyll (Chl) *a*/*b* antenna in higher plants contributes to photosynthesis by absorbing light energy and funneling it into the photosynthetic reaction centers where the conversion into an electrochemical potential takes place. The components of this photosynthetic antenna, the light-harvesting Chl *a*/*b* complexes (LHC), fulfil this task with the help of numerous protein-bound pigments, carotenoids, and Chl *a* and Chl *b*, which exchange absorbed energy rapidly and with high efficiencies.[1] The most abundant Chl *a*/*b* complex is the major LHC of photosystem II, LHCIIb, which comprises roughly 50% of the total Chl in higher plants. This complex can be reconstituted in vitro from its protein and pigment components, using either denatured thylakoid proteins[2] or recombinant LHCIIb apoprotein, Lhcb1.[3] Recombinant LHCIIb exhibits structural, biochemical, and spectroscopic properties very similar to those of native LHCIIb.[3–5]

The in vitro reconstitution of recombinant LHCIIb opens up the possibility of introduce useful modifications into the structure by altering the amino acid sequence. Thus, anchors have been generated for immobilizing the complex or for site-specific fluorescence labeling.[6] This property makes recombinant LHCIIb a promising candidate for designing hybrid biological–chemical structures that contain an ordered arrangement of fluorophores.

Herein we show that recombinant LHCIIb can be coupled to an artificial energy trap, benzoylterrylene-3,4-dicarboximide (BTI). This NIR dye collects, by efficient energy transfer, a large fraction of the light energy absorbed by the LHCIIb pigments, which makes the LHCIIb–BTI construct a simple model of a photosystem consisting of a light-harvesting pigment–protein complex and an energy trap.

To couple BTI (**1**)[7] to cysteine side chains in the protein, a maleimido derivative of the dye was constructed (Scheme 1). Maleimido BTI (**2**) was synthesized in a four step reaction




Scheme 1. R = *tert*-butylphenol; a) $Br_2$, $CHCl_3$, RT, 16 h, 78%; b) (4-pinacolylborono)-Ph$(CH_2)_2$NBOC, $Pd(PPh_3)_4$, $K_2CO_3$(aq), toluene, 110°C, 18 h, 84%; c) TFA/$CH_2Cl_2$, RT, 2 h, 93%; d) *N*-succinimidyl-4-maleimidobutyrate, $Et_3N$, $CH_2Cl_2$, RT, 6 h, 78%. BOC = *tert*-butyloxycarbonyl, TFA = trifluoroacetic acid.

from **1**. Bromination of **1** followed by Suzuki coupling with the BOC-protected (4-pinacolylborono)phenylethylamine afforded the corresponding BOC derivative of phenylethylamino BTI. After acidic removal of the BOC protecting group, the phenylethylamino BTI was transformed into **2** with *N*-succinimidyl-4-maleimidobutyrate.

Maleimido BTI (**2**) was attached to a single cysteine residue near the N terminus of the Lhcb1 mutant S3C. The site specificity of the labeling reaction was verified by the fact that another Lhcb1 mutant containing no cysteine at all (C79S) did not bind any BTI maleimide (not shown). Lhcb1–BTI was purified by preparative electrophoresis, in which it migrated more slowly than the nonlabeled protein. Subsequently the

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