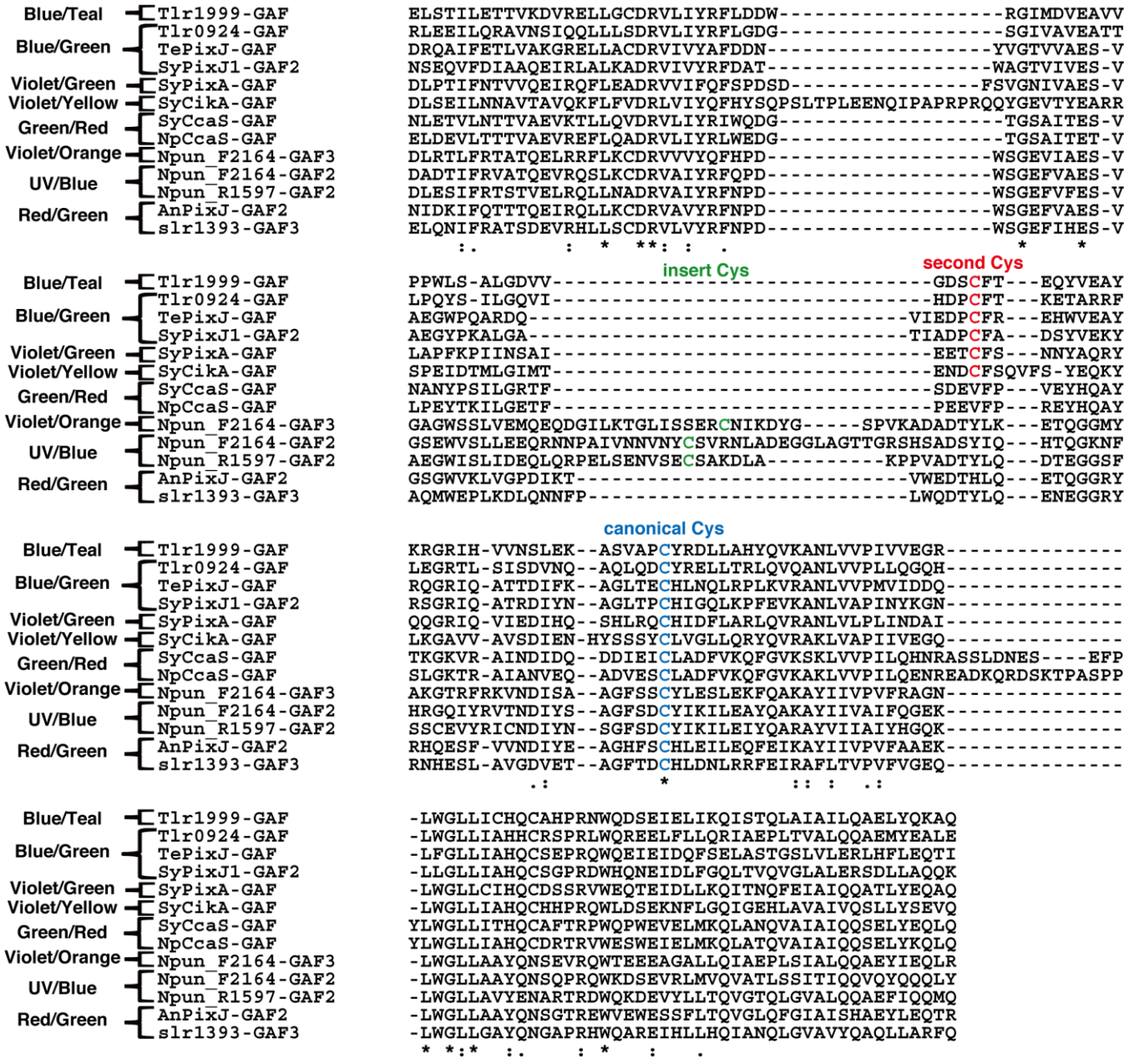

Figure S1. Sequence alignment (CLUSTALX) of the chromophore-binding GAF domains of the experimentally confirmed CBCRs.

CBCRs perceive two colors corresponding to the chromophore configuration of C15-*Z* or C15-*E*. The canonical Cys (blue) and the second Cys (red) correspond to Cys274 and Cys246 in Tlr1999, respectively. The insert Cys is highlighted in green.

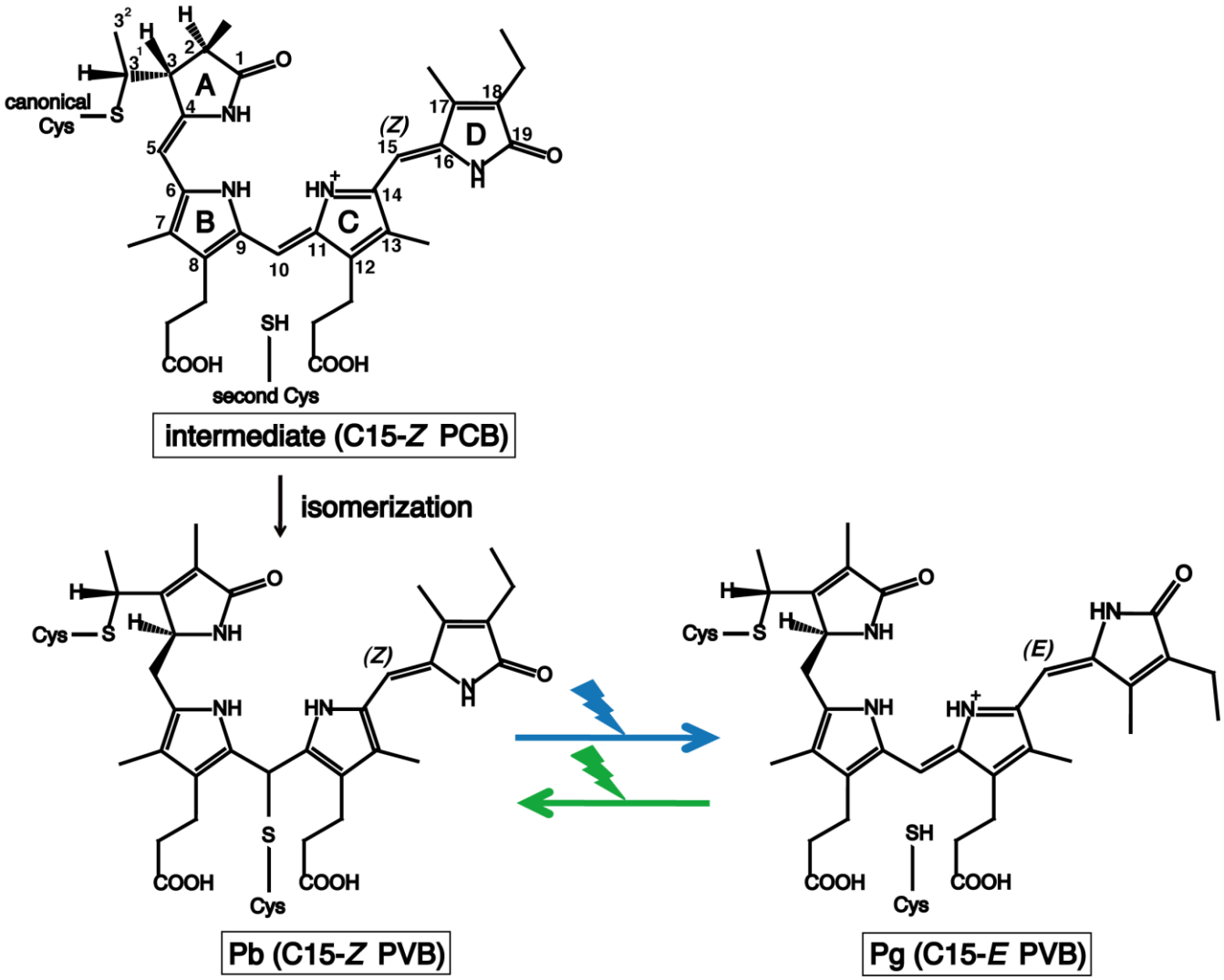

Figure S2. The chromophore structure and the reversible Cys attachment model of Pb/Pg photoconversion.

Upper left, the assembly intermediate (C15-*Z* PCB), lower left, the Pb form (C15-*Z* PVB with an additional thioether linkage between the second Cys and C10), and lower right, the Pg form (C15-*E* PVB without the additional linkage). Note that the chromophore is anchored to the canonical Cys via thioether linkage at $C3^1$ of ring A. PCB is isomerized to PVB by auto-isomerase activity.

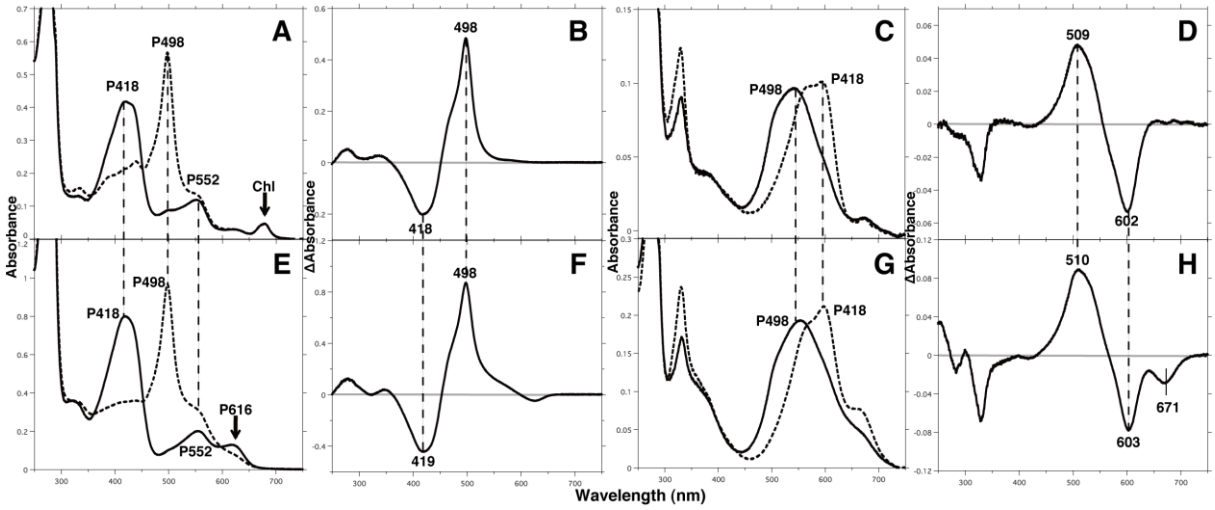

Figure S3. Absorption and difference spectra of Tlr1999-GAF purified from cyanobacterial cells (A~D) and *E. coli* (E~H).

A and E, absorption spectra of P418 (solid line) and P498 (broken line). B and F, the difference spectra of P498 minus P418. C and G, absorption spectra of denatured P498 (solid line) and denatured P418 (broken line). D and H, the difference spectra of the denatured P498 minus denatured P418. For comparison, the panel A and panel C were reproduced from Figure 1D and 1E, respectively, in the main text.

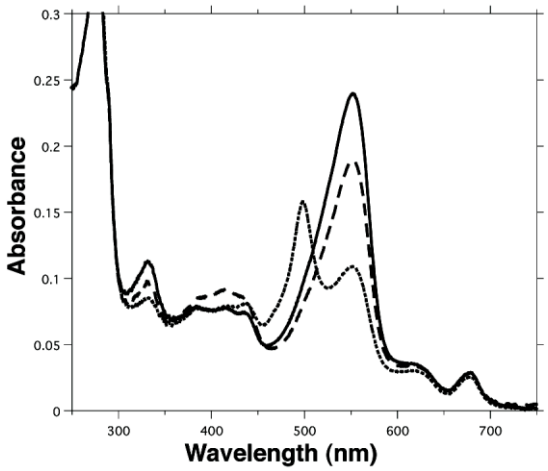

Figure S4. Treatment of IAM-modified Tlr1999-GAF with L-cysteine.

Absorption spectra. The IAM-modified Tlr1999-GAF after dialysis (solid line) was treated with 30 mM L-cysteine (broken line) and then irradiated with blue light (dotted line).

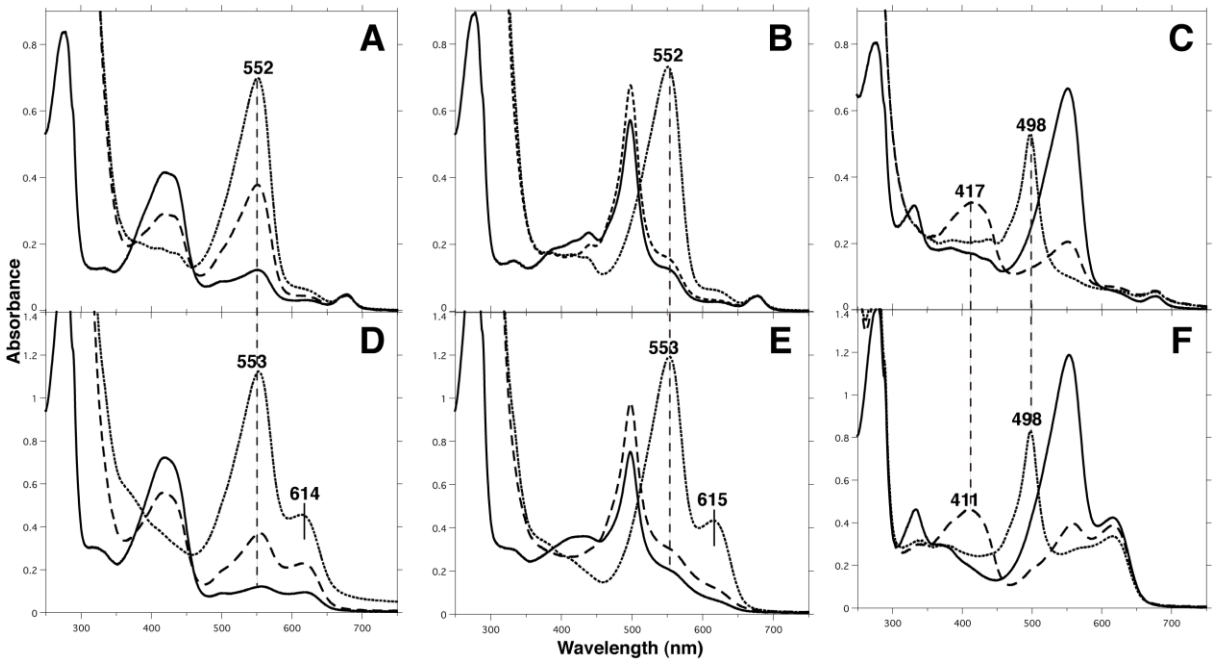

Figure S5. Effects of IAM and DTT on Tlr1999-GAF from *E. coli* in comparison with those from cyanobacterial cells.

Absorption spectra. A~C, protein prepared from cyanobacterial cells; D~F, protein prepared from *E. coli.* A and D, 0 h (solid line), 1 h (broken line), and 21 h (dotted line) after reaction of P418 with 50 mM IAM. B and E, 0 h (solid line) and 5 min (broken line) after reaction of P498 with 50 mM IAM. No further change in the absorption spectrum was observed after 5 min. After incubation for 1 h, P498 was irradiated with teal light (dotted line). C and F, dialyzed, IAM-modified Tlr1999-GAF (solid line) was treated with 20 mM DTT (broken line), irradiated with blue light (dotted line). For comparison, the panel A, panel B, and panel C were reproduced from Figure 3A, Figure 3B, and Figure 4A, respectively, in the main text.