

**Fig. S1** Verification of shade conditions and determining leaf number of Arabidopsis plant for Raman spectroscopy. **a** Measurements of petiole length and leaf blade area of wild type (Col-0) Arabidopsis plants grown in shade conditions. Bars denote average  $\pm$  SE (n=4). **b** Left panel, schematic diagram of purpose-built Raman Spectroscopy system used in this study. Right panel, photograph of the Raman Spectroscopy system. Inset shows the leaf of a plant resting on the sample holder for measurement. **c** Left panel, Raman spectrum for each leaf number. Inset focused on 1521 cm<sup>-1</sup> Raman peak. Right panel, measured peak intensities at 1521 cm<sup>-1</sup> Raman shift. Bars denote average  $\pm$  SE (n=8). **d** Development of leaf number 3 of Arabidopsis plant. Numbers represent leaf number, according to order of development. C represents cotyledon. Scale, 1cm. WL, white light; MS, moderate shade; DS, deep shade; DAG, days after germination.



**Fig. S2** Raman spectra of Arabidopsis plant under shade conditions. **a** Raman spectra of Arabidopsis leaf blades and petioles under different shade conditions. (leaf blade: n=8, petiole: n=4). **b** Three-dimensional principal component analysis (PCA) plot of wild type (Col-0) Arabidopsis under shade conditions. **c** Raman spectra for carotenoid standards. WL, white light; MS, moderate shade; DS, deep shade.



**Fig. S3** Raman spectra of same-age wild type (Col-0) Arabidopsis plants with different duration of shade treatment. Number of hours (h) or days (d) represents the duration of shade treatment. Control plants are not exposed to shade. (leaf blade: n=10, petiole: n=8).



**Fig. S4** Raman spectra of Arabidopsis plants from seedling to mature stage in time-course shade experiment. **a** Phenotype of plants at different age and different durations of shade treatment. 10 d-old plants were subjected to 7 d treatment of white light (WL) or deep shade (DS). **b** Carotenoids Raman peak of plants in **a**. Dashed line indicates difference in peak intensity between WL and DS. DAG, days after germination.



**Fig. S5** Raman spectra of Arabidopsis phytochrome mutants under shade conditions. **a** Petiole length and leaf blade area of *phyB-9<sup>BC</sup>* and *phyA-211* under shade. Bars denote average  $\pm$  SE (n=3). **b** Raman spectra of leaf blades and petioles of *phyB-9<sup>BC</sup>* and *phyA-211* in **a** (leaf blade: n=8, petiole: n=4). WL, white light; MS, moderate shade; DS, deep shade.

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**Fig. S6** Raman spectra of wild type (Col-0) Arabidopsis leaf blades and petioles in low to high density planting. (leaf blade: n=5, petiole: n=3).



Fig. S7 Raman spectra of leafy vegetables under shade conditions. a Development of leaf number 1 (blue arrowhead) in vegetables. Scale, 2 cm. b Measurements of petiole length, leaf blade area, hypocotyl length, and epicotyl length of Kai Lan and Choy Sum grown in shade conditions. Bars denote average  $\pm$  SE (n=3). c Raman spectra of leaf blades and petioles of Kai Lan and Choy Sum in a (leaf blade: n=8, petiole: n=4). WL, white light; MS, moderate shade; DS, deep shade; C, cotyledon.



**Fig. S8** Raman spectra of leafy vegetables with different duration of shade treatment. **a** Raman spectra of leaf blades and petioles of Kai Lan (n=5). **b** Raman spectra of leaf blades and petioles of Choy Sum (n=4).



**Fig. S9** Raman spectra of leafy vegetables in high density planting. **a** Measurements of petiole length, leaf blade area, hypocotyl length, and epicotyl length of Kai Lan and Choy Sum grown in low to high density conditions. Bars denote average  $\pm$  SE (n=4). **b** Raman spectra of leaf blades and petioles of Kai Lan and Choy Sum in **a** (leaf blade: n=6, petiole: n=4).



**Fig. S10** Raman spectra of various plant species under shade conditions. (Baby Bok Choy: leaf blade n=7, petiole n=4; Bok Choy 'Purple King': leaf blade n=6, petiole n=6; Romaine Lettuce: leaf blade n=6, petiole n=6; *Nicotiana benthamiana*: leaf blade n=3, petiole n=3; *Nicotiana tabacum*: leaf blade n=6, petiole n=3). WL, white light; MS, moderate shade; DS, deep shade.