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Isoxazolone-based Single Crystals with Large Second Harmonic Generation Effect

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Synthesis

(Z)-4-(4-(Dimethylamino)benzylidene)-3-phenylisoxazol-5(4H)-one (DLS): DLS was synthesized according to the literature. The product was found to be pure and no further purification was necessary.

(Z)-4-(4-methoxybenzylidene)-3-phenylisoxazol-5(4H)-one (MLS): A mixture of 20 mmol ethyl benzoylacetate, 20 mmol hydroxylamine hydrochloride，and 11 mmol DABCO was refluxed in 50 mL ethanol for 3 min, after then 20 mmol p-methoxybenzaldehyde was added and the mixture was further refluxed for 8h. After completion of the reaction, the mixture was cooled to room temperature and the crystal product was collected by filtration. The product was found to be pure and no further purification was necessary.

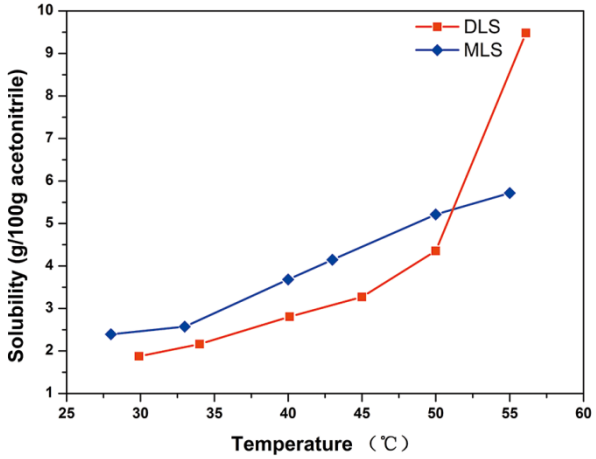

Fig. S1 Solubility of DLS and MLS in acetronitrile.

UV-vis Diffuse Reflectance Spectroscopy.

Fig. S2 clearly demonstrates that the short cut-off wavelengths of DLS and MLS crystals are located at 561 nm and 472 nm, respectively, corresponding to the energy band gaps of 2.21 eV and 2.63 eV as obtained by the straightforward extrapolation method.[1] The cut off wavelengths are consistent with transmission spectra results.

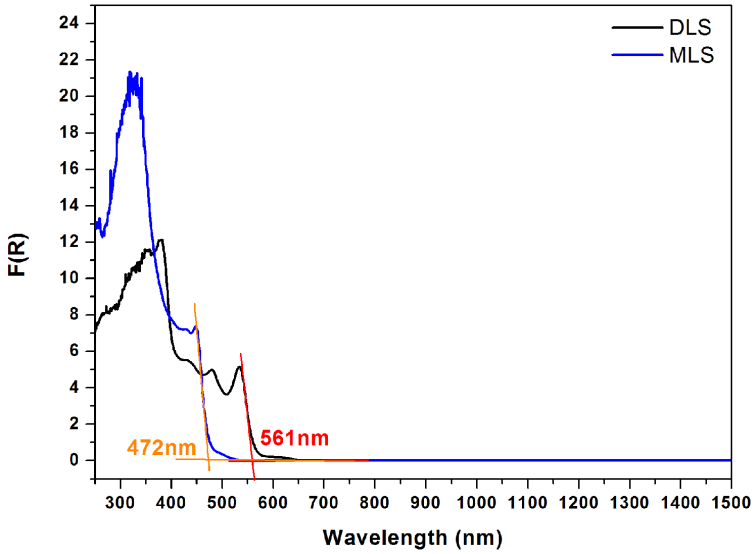

Fig. S2 UV-vis-NIR diffuse reflectance spectra of DLS and MLS crystals.

Table S1 The calculated first-order hyperpolarizability β(a.u.) of DLS and MLS in B3LYP/6-311G* using Gaussian03 program.

	DLS	MLS
β_{111}	334.9	-213.27
β_{112}	-25.5	27.5
β_{113}	14.1	-21.9
β_{122}	23 .1	-8.1
β_{123}	146.2	-84.9
β_{133}	173.6	-29.8
β_{222}	-136.7	13.0
β_{223}	-706.5	835.7
β_{233}	931.1	9.2
β_{333}	-4592.6	2917.9

Table S2 Comparison of the SHG coefficients and SHG effect of the DAST, DSTMS, OH1, DLS, and MLS.[2-3]

	DLS	MLS	OH1	DAST	DSTMS
SHG coefficients (pm/V)	-	-	d_{33} = 120±10	d_{11} = 210±55	d_{11} = 214±20
Powder SHG effect	2.8	1.5	1	3	3

References

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