Among the occurrence of SH, a maximum signal power of $5$ nW can be obtained via the signal fiber. In comparison of the collecting efficiency of the two fibers, the fiber-cavity coupling is optimized so that the SH signal from the pump fiber is observable but the maximum power is over one order of magnitude weaker than the power from the signal fiber. From either fibers, SH signal is absent when the pump is off-resonance with cavity modes, which helps to eliminate the possibility of spurious signals such as the second order diffraction of the EMCCD grating.

Second-order nonlinear optical processes lie in

the heart of many applications in both classical and quantum regime, e.g. frequency conver-

sion, quantum squeezing and entanglement. In-

version symmetry inhibits the second-order non-

linear dipole response in centrosymmetric and

amorphous materials. Symmetry breaking at sur-

faces/interfaces can produce second-order nonlin-

earity, but its signal requires high-power excita-

tion and is hard to be distinguished from the con-

tribution of bulk multipole nonlinearity. Here, for

the first time, we report second harmonic genera-

tion (SHG) originating deterministically from the

surface nonlinearity of a silica microcavity. The

bulk multipole nonlinear effects are eliminated

via pumping a fundamental mode with transverse

electric polarization, enabling the identification

of surface nonlinear response. The doubly res-

onant enhancement of ultrahigh-Q modes lowers

the pump power below one milliwatt, and boosts

the SHG conversion efficiency to 0.049% W−1 , ex-

ceeding 10 orders of magnitude compared with the

non-resonance case. This work can trigger appli-

cations in cavity-sensing with surface specificity,

extend the frequency conversion range of silica

photonic devices and possibly push the nonlinear-

ity down to the quantum regime.