Electronic Supplementary Information (ESI)

When Au NPs with different sizes have the same particle number, they will have different surface areas and different mass. Similarly, when the Au NPs with different sizes have the same surface area or mass, they will have different particle numbers and different mass or different surface areas. Table S1 shows the comparison of all the particles used in our study when we examine the particle number, surface area, and mass effects.

Table S1 Comparison of NP samples studied in this research in particle number, surface area, and mass.

Diameter (nm)	Particle Number (PN) Study			Surface Area (SA) Study			Mass (MA) Study		
	PN	SA	MA	PN	SA	MA	PN	SA	MA
5	3.17×10^{8}	2.49×10^{-4}	4.00×10^{-10}	3.17×10^{10}	2.49×10^{-2}	4.00×10^{-8}	3.17×10^{11}	2.49×10^{-1}	4.00×10^{-7}
20	3.17×10^{8}	3.99×10^{-3}	2.56×10^{-8}	1.98×10^{9}	2.49×10^{-2}	1.60×10^{-7}	4.95×10^{9}	6.22×10^{-2}	4.00×10^{-7}
50	3.17×10^{8}	2.49×10^{-2}	4.00×10^{-7}	3.17×10^{8}	2.49×10^{-2}	4.00×10^{-7}	3.17×10^{8}	2.49×10^{-2}	4.00×10^{-7}
100	3.17×10^{8}	9.96×10^{-2}	3.20×10^{-6}	7.92×10^{7}	2.49×10^{-2}	8.00×10^{-7}	3.96×10^{7}	1.24×10^{-2}	4.00×10^{-7}

Note: The units for PN, SA and MA are particle/mL, cm^2/mL, and g/mL respectively.

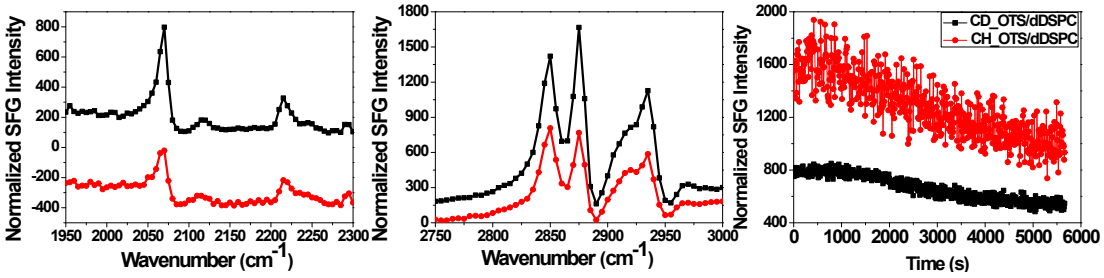

Figure S1. SFG spectra in the (a) C-D and (b) C-H stretching frequency ranges before (black circle) and after (red circle) the introduction of Au-BBI-20 for OTS/dDSPC bilayer respectively. (c) Time dependent SFG signal in C-D (black) and C-H (red) stretching frequency range for Au-BBI-20.

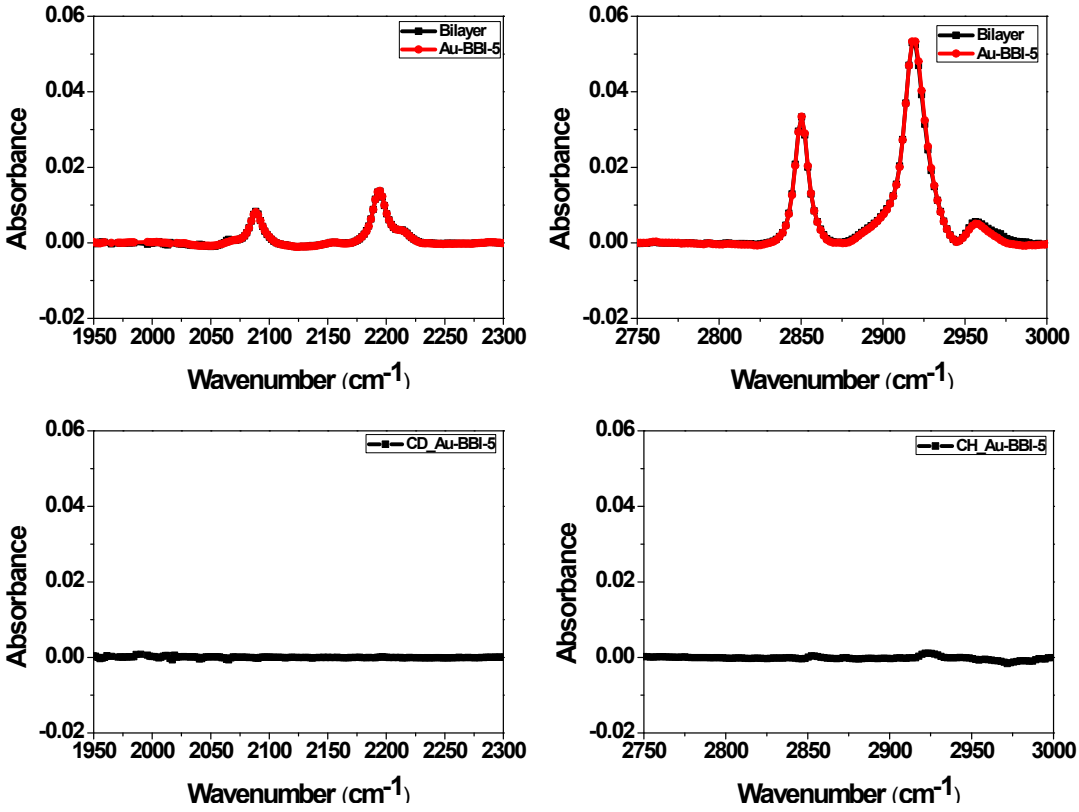

Figure S2. ATR-FTIR spectra in (a) C-D and (b) C-H stretching frequency ranges from the dDSPC/DSPC bilayer before (-●-) and after (-●-) the addition of 5 nm Au NPs to the bilayer subphase. The difference ATR-FTIR signals shown in (c) C-D and (d) C-H stretching frequency ranges are obtained by subtracting the black signals from the red signals shown in (a) and (b).

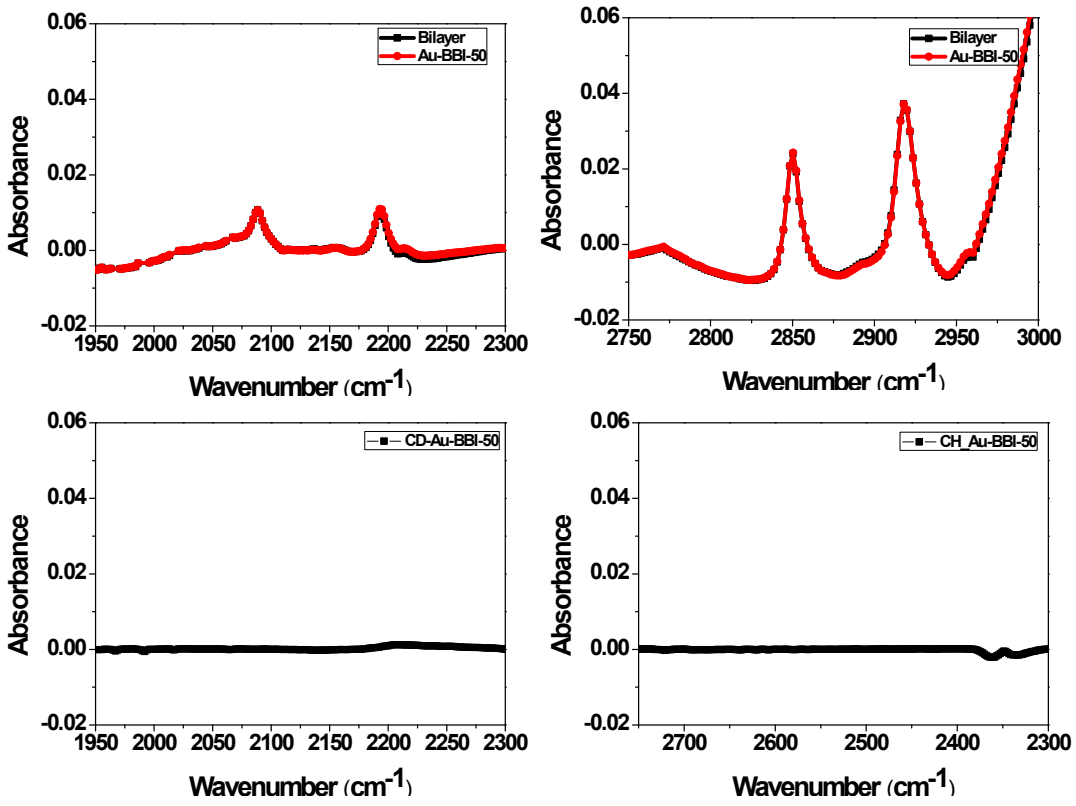

Figure S3. ATR-FTIR spectra in (a) C-D and (b) C-H stretching frequency ranges from the dDSPC/DSPC bilayer before (-●-) and after ((-●-) the addition of 50 nm Au NPs to the bilayer subphase. The difference ATR-FTIR signals shown in (c) C-D and (d) C-H stretching frequency ranges are obtained by subtracting the black signals from the red signals shown in (a) and (b).

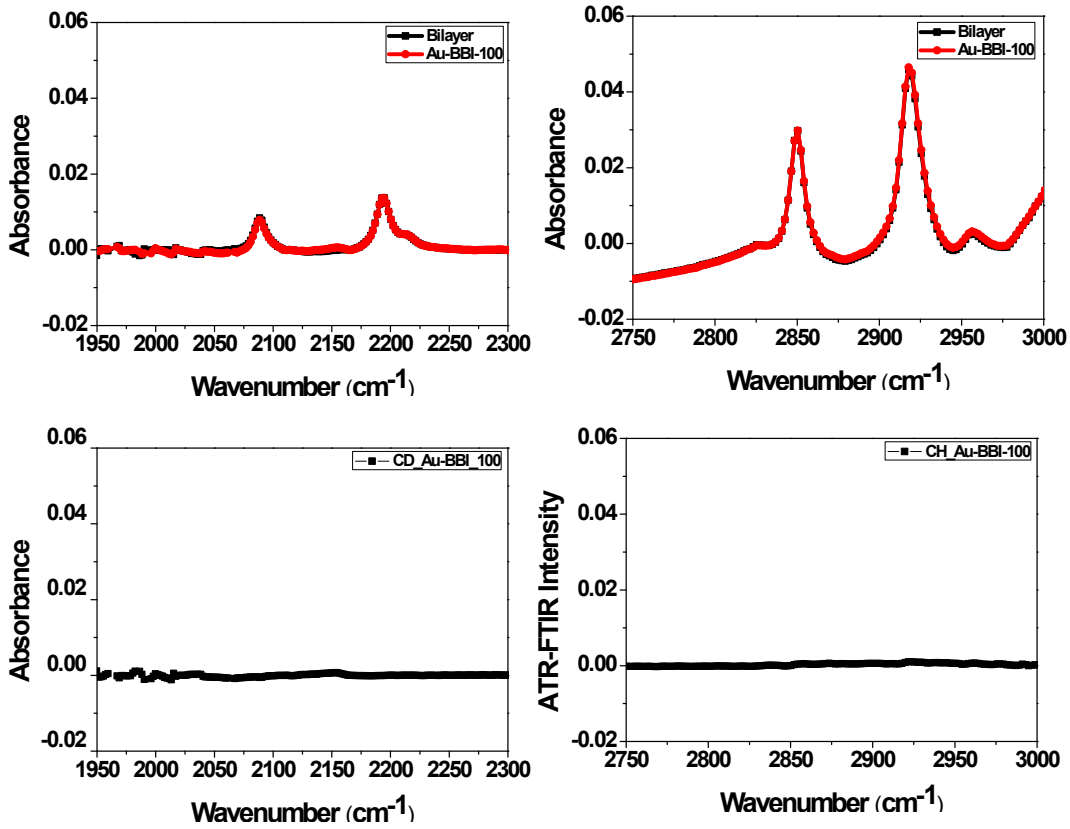

Figure S4. ATR-FTIR spectra in (a) C-D and (b) C-H stretching frequency ranges from the dDSPC/DSPC bilayer before (-●-) and after ((-●-) the addition of 100 nm Au NPs to the bilayer subphase. The difference ATR-FTIR signals shown in (c) C-D and (d) C-H stretching frequency ranges are obtained by subtracting the black signals from the red signals shown in (a) and (b).

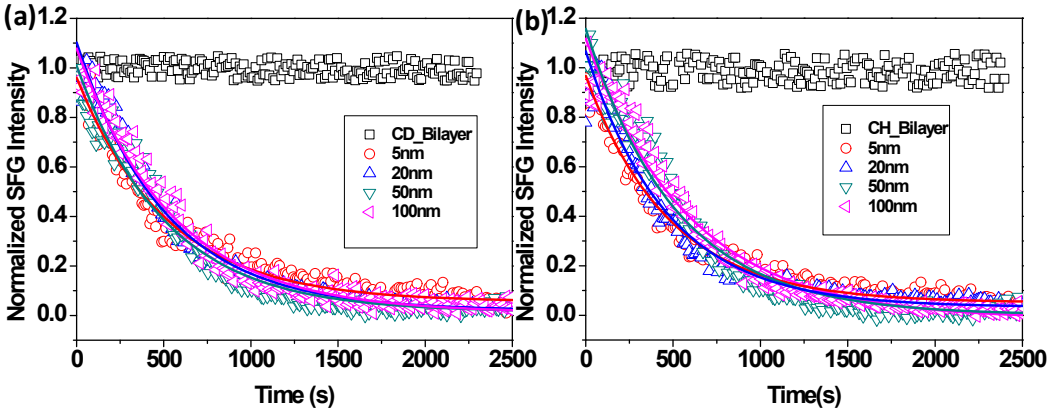

Figure S5. Time-dependent SFG signal intensity changes in (a) C-D and (b) C-H stretching frequency ranges when Au NPs (5, 20, 50, and 100nm) interacting with the dDSPC/DSPC bilayer at 2.00 x10^{-7} g/mL. The dots are experimental data and the lines are fitting results.