To compare our method with effective medium approximation (EMA) as a traditional effective medium method, we calculated the SPR profile by EMA. The effective dielectric constant, ε, by EMA is defined as Eq. (1S),

$$\sigma \frac{\varepsilon_i - \varepsilon}{\varepsilon_i + 2\varepsilon} + (1-\sigma)\frac{\varepsilon_a - \varepsilon}{\varepsilon_a + 2\varepsilon} = 0 \qquad (1S)$$

where ε_i represents the dielectric constant of the material which has coverage, σ, and ε_a is a dielectric constant of the other material.

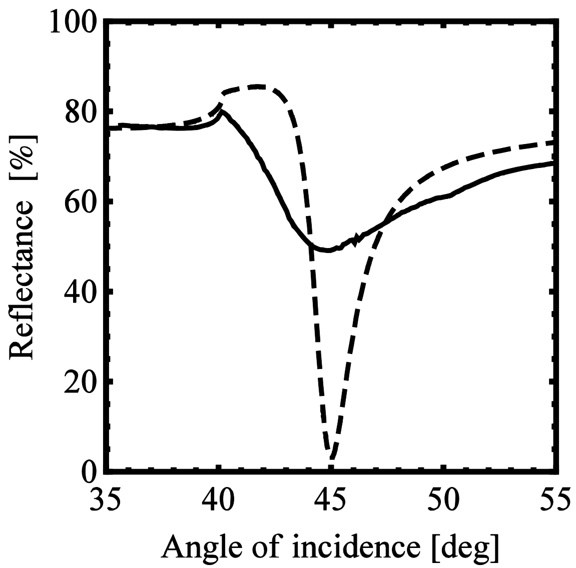

Figure 1S SPR profiles measured (solid line) and calculated by the effective medium approximation (broken line) for sample III.

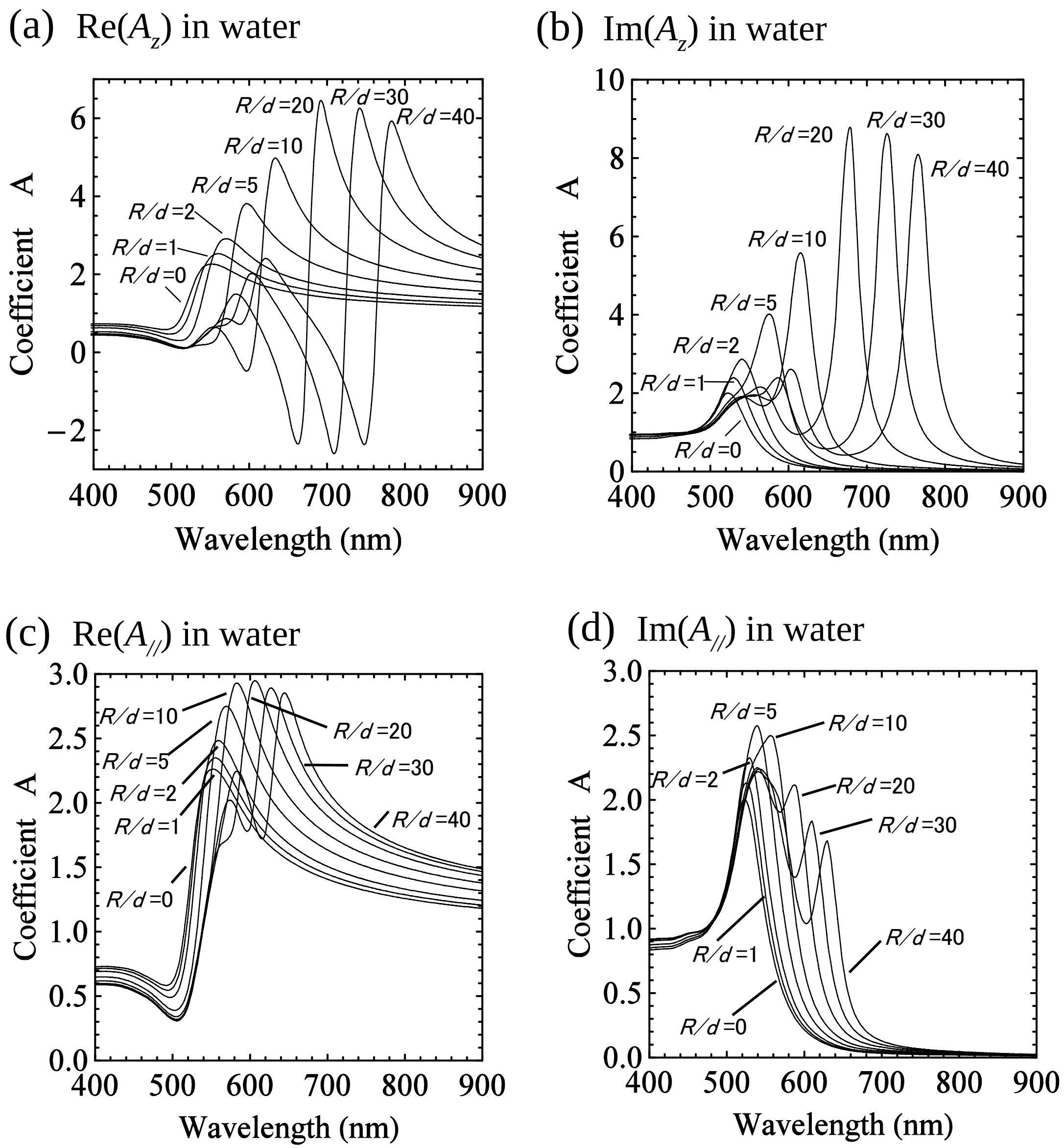

Figure 2S The parameter A_z and $A_{//}$ calculated using Eq. (3) and (4) at different *R/d* ratios in water. (a) Real part of A_z . (b) Imaginary part of A_z . (c) Real part of $A_{//}$. (d) Imaginary part of $A_{//}$.

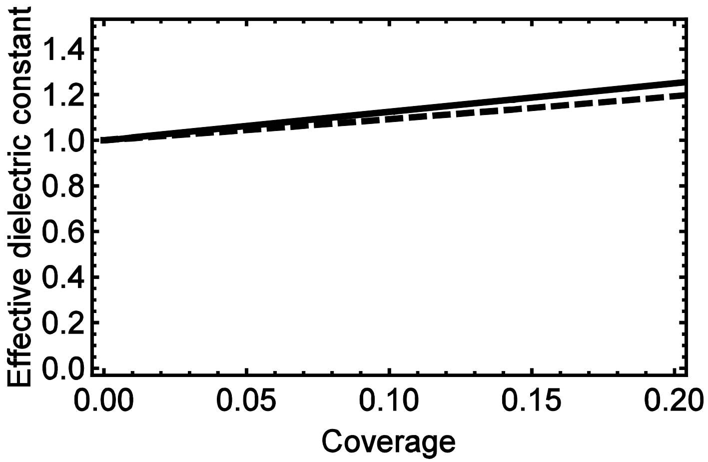

Figure 3S Effective dielectric constants of the gap-supporting layer against the coverage calculated by Eq.(30) (solid line) and calculated by the EMA, Eq. (1S) (broken line) . Since the error of dielectric constant between two methods was 5% at σ=0.2, Eq.(30) was employed for simplicity.

supplement 4

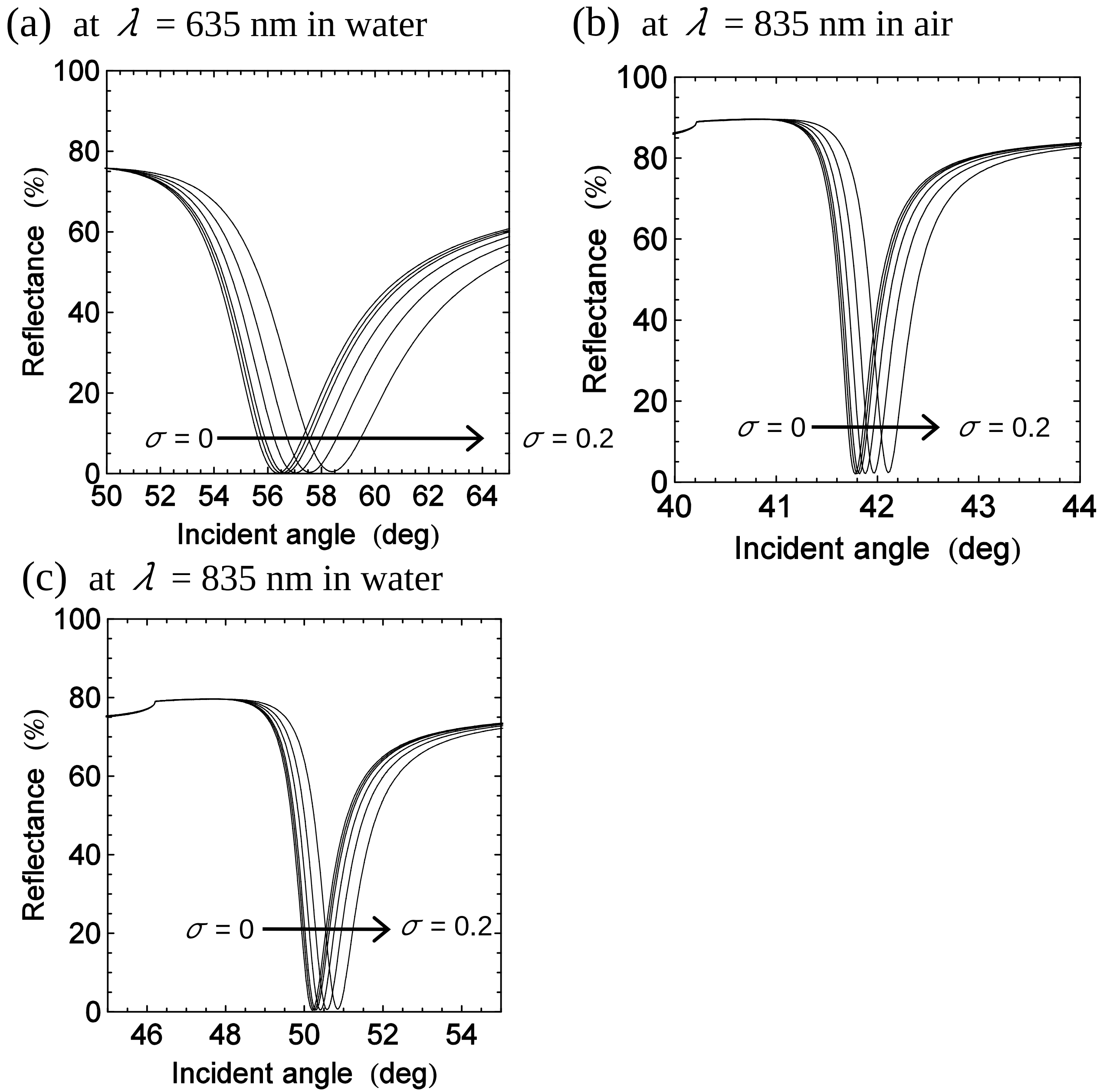

Figure 4S Calculated SPR profiles at different coverages σ = 0, 0.01, 0.02, 0.05, 0.1 and 0.2 at λ = 635 nm in water (a), λ = 835 nm in air (b) and λ = 835 nm in water (c). In the case of water, a rectangular prism of high index grass (refractive index 1.86) was assumed.

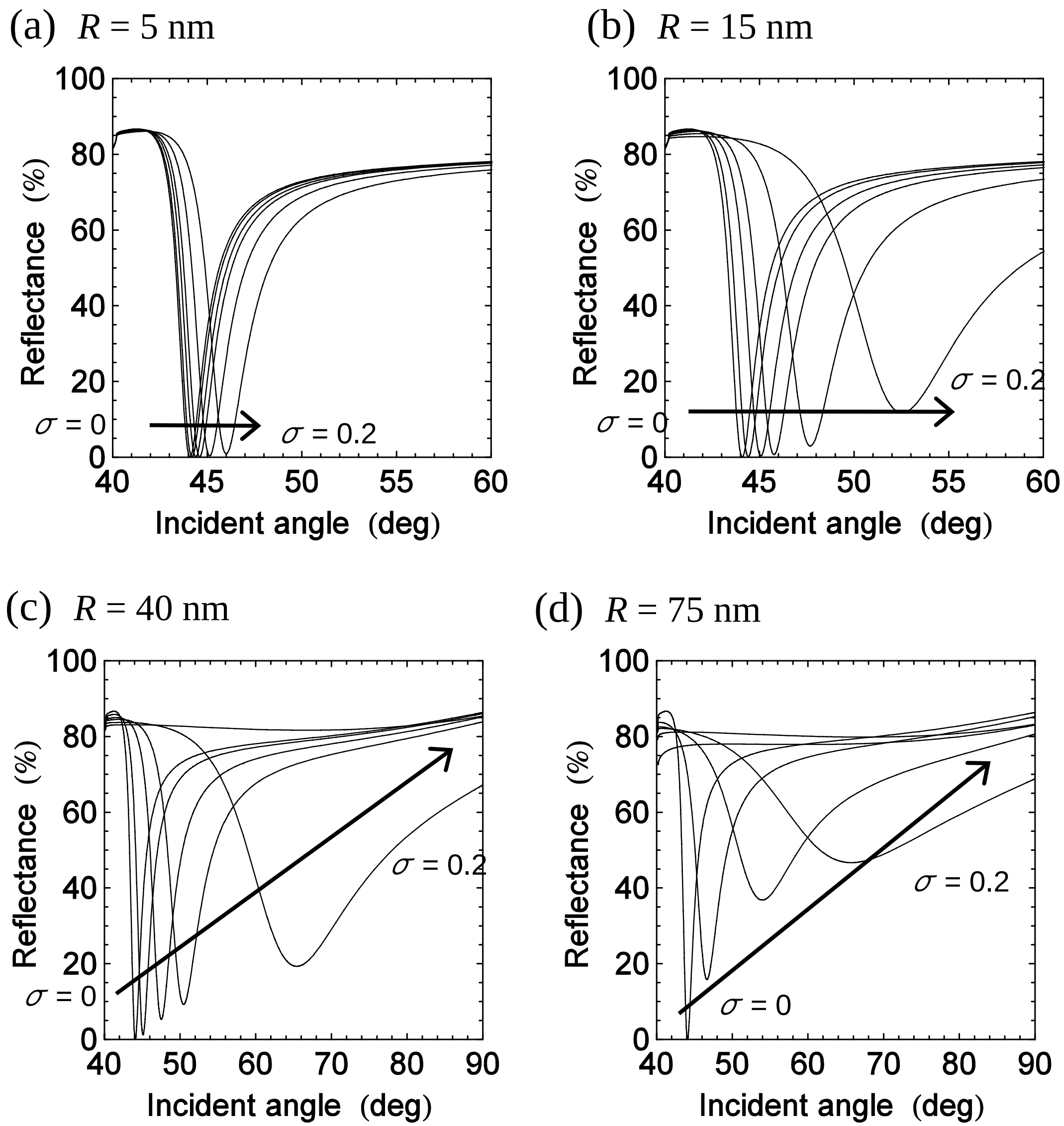

Figure 5S Calculated SPR profiles at different coverages σ = 0, 0.01, 0.03, 0.05, 0.1 and 0.2 at λ = 635 nm in air. The thickness of the gap-supporting layer d = 5 nm. (a) R = 5 nm , (b) R =15 nm, (c) R = 40 nm and (d) R =75. In the case of water, a rectangular prism of high index grass (refractive index 1.86) was assumed.