arrow in Fig 4(a)), we obtain calculated line profiles, similar to the experimental ones shown in Fig. 2(b) and Fig. 3(a). From the calculated profiles we determine the FWHMs of the gold disks. The results of our simulations are summarized in Fig. 4(b). We find that the FWHMs of the peaks are reduced at smaller tapping amplitudes and higher harmonics, therefore *confirming the trends* obtained in the experiment.

We note that the CDM model is widely used to verify overall trends in s-SNOM experiments, providing *qualitatively correct* results [1,19,25–30] Quantitative differences between the model and our experiment we attribute to the finite sizes of the tip and the disks, which are not taken into account in the model. In particular, the near-field probe employed in the experiment is a large (several micrometers long) metallized tip. The polarization developed in such tip under the external illumination and due to the near-field interaction with the sample can differ significantly from that of the small sphere representing the tip in the model. Although, a quantitative treatment of s-SNOM contrasts is possible with the finite-dipole model, where the tip is treated as a spheroid [31], this model is only applicable in the case of homogeneous samples [31] or for small spherical nanoparticles located directly below the tip [32]. For describing subsurface near-field contrasts in more complex samples, such as the ones studied in this work, future improved models have to be developed.

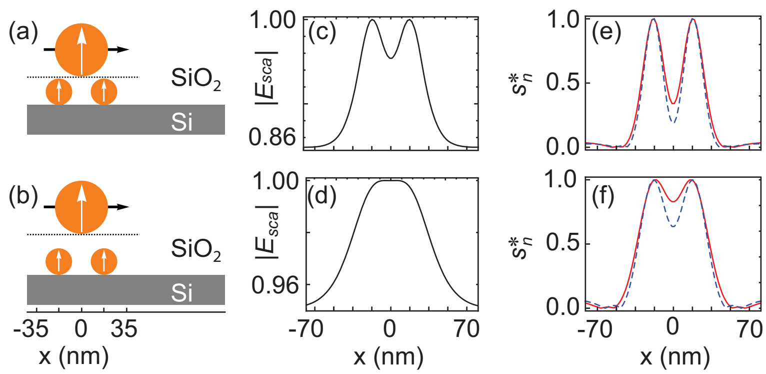

Fig. 5. CDM simulations. (a, b) Schematics of the simulation showing the probe sphere (tip, radius 20 nm) scanning across two small spheres (radii 10 nm). (c, d) Calculated near-field profile E_{sca}. (e, f) Calculated near-field profiles of 3rd (solid red line) and 4th (dashed blue line) harmonics. The top panels show the simulation with the spheres buried right below the SiO_2 surface. The bottom panels show the simulation with the spheres buried at the depth of 15 nm beneath it. All signals are normalized to their own maxima.

We have further utilized the CDM model to simulate the near-field signal E_{sca} and its harmonics s_n as the tip, described as a sphere dipole of 20 nm in radius, scans along a sample consisting of a pair of small Au nano-spheres buried below the SiO_2 surface (Fig. 5). The small spheres are 10 nm in radius and separated by a gap of 15 nm. When the spheres are located right below the tip (a), they can be resolved in the E_{sca} profile (c) and in s_n (e). The situation changes dramatically when the spheres are buried at 15 nm depth (b). The near-field signal E_{sca} exhibits a single peak not revealing the presence of two spheres (d). In contrast, the higher harmonic profiles s_3^* and s_4^* clearly resolve the two spheres (f), which provides a further theoretical support for the enhancement of the resolution by higher harmonic demodulation.

In conclusion, we have demonstrated that the spatial resolution and depth contrast in subsurface s-SNOM can be enhanced by operating at small tapping amplitudes and by demodulating the detector signal at higher harmonics of the tapping frequency. This improvement comes at

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the cost of signal strength. Our simulations qualitatively corroborate these experimental findings and indicate that the resolution improvement becomes more significant when smaller, more closely spaced subsurface objects are imaged.

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