Supporting Information

Effect of Dipole Coupling on Near-IR LSPR and Coherent Phonon Vibration of Periodic Gold Pair Nanocuboids

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Table S1 The oscillation periods of the decay profiles at the bleaching (1000 or 900 nm) and absorption wavelengths (1100 or 940 nm) and bleaching peaks (λbleach) for +45° and -45° probe polarizations with the fitting function of

\[ R(t) = A_{c-p} \exp(-t/τ_{c-p}) + \sum_{i=1,2,3} A_i \exp(-t/τ_i) \cos(2πt/T_i + φ_i) + A_\text{bleach} \exp(-t/τ_\text{bleach}). \]

<table>
<thead>
<tr>
<th>Probe Polarization</th>
<th>T1 (ps)</th>
<th>T2 (ps)</th>
<th>T3 (ps)</th>
</tr>
</thead>
<tbody>
<tr>
<td>+45°</td>
<td>1000</td>
<td>85</td>
<td>126</td>
</tr>
<tr>
<td></td>
<td>1100</td>
<td>85</td>
<td>126</td>
</tr>
<tr>
<td>λbleach</td>
<td></td>
<td>86</td>
<td>126</td>
</tr>
<tr>
<td>-45°</td>
<td>900</td>
<td>90</td>
<td>107</td>
</tr>
<tr>
<td></td>
<td>940</td>
<td>89</td>
<td>103</td>
</tr>
<tr>
<td>λbleach</td>
<td></td>
<td>91</td>
<td>105</td>
</tr>
</tbody>
</table>

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Figure S1 The Fourier transforms for the modulated portion of the decay profiles at the bleaching peaks (+45°) and absorption peak (0-nm, -45°) of S600 with various nanogaps.
Figure S2 The decay profiles from 960 nm to 1150 nm were fitted with two damped cosine functions by convenient for the array with 600 nm separation and 0 nm nanogap. All of the experimental data and the fitting results for array are illustrated in the bellowing contour with colors and solid lines, respectively. The fitting function is written in the form

\[
R(t) = A_{e-p} \exp(-t / \tau_{e-p}) + \sum_{\nu=1,2} A_{\nu} \exp(-t / \tau_{\nu}) \cos(2\pi t / T_{\nu} + \varphi_{\nu}) + A_{\text{th}} \exp(-t / \tau_{\text{th}}). 
\]
Figure S3 The comparison of the oscillation amplitudes of bleaching peaks between two arrays with different nanogaps (+45° polarized probe pulses), by using same scale of y-axes for comparing the amplitudes (a) and different scale of y-axes for comparing the damping constants (b). The samples are the gold pair nanocuboid arrays with 0-nm or 10.6-nm nanogaps within one pair and 600-nm separation between pairs. The average damping constants of the modulated portion of bleach peak for the seven arrays (c).
Figure S4 The comparison of the oscillation amplitudes of bleaching peaks with the $+45^\circ$ and $-45^\circ$ polarized probe pulses. The sample is the gold pair nanocuboid array with a 0-nm nanogap within one pair and 600-nm separation between pairs.
Figure S5 Decay profiles and the fitting curves for G0nm at the absorption wavelength for the array with 400 nm separation (a) and the bleaching wavelengths for the arrays with various separations from 450 nm to 700 nm. (b) The oscillation of bleaching peaks of G0nm with various separations of 500, 650 and 700 nm. The central bleaching peaks of 400-nm and 450-nm separation arrays are beyond our detection region. The bleaching peaks for the separation of 500, 550 and 600 nm are showing the similar position and only the results for 500-nm separation arrays are shown here.
Figure S6 Decay profiles at the bleaching wavelengths at 890, 900, 960 and 1050 nm (a) and oscillation of bleaching peaks (b) for G5nm with the separations of 400, 500, 600 and 700 nm.
Figure S7 Decay profiles at the absorption wavelengths (a) and oscillation of bleaching or absorption peaks (b) for G10nm with various separations. The bleaching peaks of the 400-nm separation array are beyond our detection region and the absorption decay profiles are illustrated for all the arrays.
Figure S8 Correlation between the nanogaps and the oscillation amplitudes of bleach peaks for all of the periodic gold pair nanocuboids (+45° probe polarization): Type I for near-field coupling (a) and Type II for far-field interaction (b).
Figure S9 FDTD simulation for the single (a) and the pair nanocuboids (b-e) with ± 1 nm structure deformation. The models for the cuboids are expanded and compressed at the edge and the tip together and the tips are modified with 1 nm radius sectors.