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| Title: | Boosting Self-interaction of Molecular Vibrations under Ultrastrong Coupling Condition |
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| Keywords: | Resonance structures<br>Quantum mechanics<br>Oscillation |
| Issue Date: | 2021 |
| Publisher: | ACS Publications |
| Citation: | The Journal of Physical Chemistry Letters, 12(17), 4313–4318. |
| Abstract: | In this letter, we investigated the modification of the oscillator strength of an asymmetric stretching band of CS2 by strong coupling to an infrared cavity photon. This is achieved by placing liquid CS2 in a Fabry–Perot resonator and tuning the cavity mode position to match the molecular vibrational transition. Ultrastrong coupling leads to an increase in the effective oscillator strength of the asymmetric stretching band of CS2. We proved this experimentally by taking the area ratio of the asymmetric stretching and combination bands by selectively coupling the former. A nonlinear increase in the oscillator strength of the asymmetric stretching band is observed upon varying the coupling strength. This is explained by a quantum mechanical model that predicts quadratic behavior under ultrastrong coupling conditions. These findings will set up a new paradigm for understanding chemical reaction modifications by vacuum field coupling. |
| Description: | Only IISERM authors are available in the record |
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| Appears in Collections: | Research Articles (/jspui/handle/123456789/9) |

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