Thermalization rate of polaritons in strongly-coupled molecular systems

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Polariton thermalization is a key process in achieving light–matter Bose–Einstein condensation, spanning from solid-state semiconductor microcavities at cryogenic temperatures to surface plasmon nanocavities with molecules at room temperature. Originated from the matter component of polariton states, the microscopic mechanisms of thermalization are closely tied to specific material properties. Here, we present the results of our recent work [1], where we investigate polariton thermalization in strongly-coupled molecular systems. We develop a microscopic theory addressing polariton thermalization through electron-phonon interactions (known as exciton-vibration coupling) with low-energy molecular vibrations. This theory presents a simple analytical method to calculate the temperature-dependent polariton thermalization rate, utilizing experimentally accessible spectral properties of bare molecules, such as the Stokes shift and temperature-dependent linewidth of photoluminescence, in conjunction with well-known parameters of optical cavities. Our findings demonstrate qualitative agreement with recent experimental reports of nonequilibrium polariton condensation in both ground and excited states, and explain the thermalization bottleneck effect observed at low temperatures. This study showcases the significance of vibrational degrees of freedom in polariton condensation and offers practical guidance for future experiments, including the selection of suitable material systems and cavity designs.

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References

[1] E.A. Tereshchenkov, I.V. Panyukov, M. Misko, V.Yu. Shishkov, E.S. Andrianov, and A.V. Zasedatelev, *Nanophotonics* 13, 14 (2024).

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