Multiple Rabi Splitting under Ultra-strong Vibrational Coupling

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Strong light-matter interaction has much potential for modifying the chemical and physical properties of molecular materials. [1] Recently we have shown for the first time that molecular vibrational transitions can be coherently coupled to vacuum states in the infrared region. [2] Here, we demonstrate that molecular liquids with very high vibrational dipolar strength can undergo multiple coupling with IR cavity modes reaching ultra-strong coupling limit (Rabi splitting >25 % of the fundamental transition energy of a vibrational band). The dynamics of the multiple polaritonic states involves both the contributions of anti-resonant term in the interaction energy and dipolar self-energy of the molecular vibrations. [3] This results in very interesting properties like polaritonic band gap opening and also a whole set of vibrational ladder of heavy polaritonic states as shown in ***Figure 1***, which should have important consequence on the reactivity energy landscape.

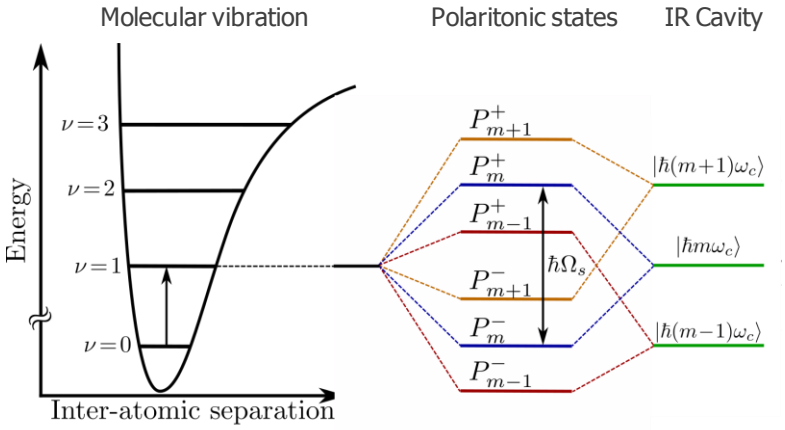

Figure 1. Schematic illustration of a vibrational transition strongly coupled to mth mode of a Fabry-Perot cavity in the strong absorbing conditions leading to multiple Rabi splitting.

References:

[1] T. W. Ebbesen *Acc. Chem. Res.*, **2016**, *49*, 2403–2412.
[2] A. Shalabney et al, *Nature Commun.*, **2015**, *6*, 5981.
[3] J. George et al, *Phys. Rev. Lett.*, **2016**, *117*, 153601.