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ABSTRACT

We extend our recently proposed theoretical framework for estimating cavity-modified equilibrium Fermi's golden rule (FGR) rate constants beyond the single cavity mode case to cases where the molecular system is coupled to multiple cavity modes. We show that the cumulative effect of simultaneous coupling to multiple modes can enhance FGR rate constants by orders of magnitude relative to the single mode case. We also present an analysis of the conditions necessary for maximizing this effect in the Marcus limit of FGR-based rate theory.

I. INTRODUCTION

This communication follows up on and extends our recently proposed general-purpose theoretical framework for estimating the effect of placing molecular matter in a photonic cavity on the rates of electronic energy and charge transfer processes.[1] Our strategy is based on equilibrium Fermi's Golden Rule (FGR) rate theory, which treats the coupling between the electronic and photonic degrees of freedom (DOF) as a small perturbation within the framework of second order perturbation theory. The reader is referred to Ref. 1 for a detailed description and analysis of our framework in the case where the photonic DOF correspond to a single cavity mode. In this communication we examine the effect of going from a model that accounts for a single cavity mode in a one-dimensional cavity to a more realistic model that accounts for multiple cavity modes in a two-dimensional cavity.

The main findings reported in this communication are: (1) Similar to the single-mode case, cavity-modified FGR rate constants can be obtained from cavity-free inputs in the case of multiple modes; (2) The cumulative effect of coupling to multiple modes can enhance FGR rate constants by orders of magnitude relative to the single mode case; (3) A generalization of the cavity-modified Marcus rate theory from the case of a single mode to the case of multiple modes.

II. HAMILTONIAN

We begin, as in Ref. 1, by considering a donor-acceptor molecular system inside a cavity, whose Hamiltonian in its most general form is given by:

$$\hat{H} = \hat{H}_{\mathrm{D}}^{\mathrm{np}}|\mathrm{D}\rangle\langle\mathrm{D}| + \hat{H}_{\mathrm{A}}^{\mathrm{np}}|\mathrm{A}\rangle\langle\mathrm{A}| + \hat{V}_{\mathrm{DA}}^{\mathrm{np}}[|\mathrm{D}\rangle\langle\mathrm{A}| + |\mathrm{A}\rangle\langle\mathrm{D}|]. \quad (1)$$

Here, $|\mathrm{D}\rangle$ and $|\mathrm{A}\rangle$ are the diabatic donor and acceptor electronic states, respectively; $\hat{H}_{\mathrm{D}}^{\mathrm{np}}$ ($\hat{H}_{\mathrm{A}}^{\mathrm{np}}$) is the Hamiltonian of the nuclear and photonic DOF when the system is in the donor (acceptor) state; and $\hat{V}_{\mathrm{DA}}^{\mathrm{np}}$ is the coupling between the donor and acceptor states (an operator in the nuclear + photonic Hilbert space in the most general case).

Assuming that the cavity modes are coupled to the electronic DOF but uncoupled from the nuclear DOF, $\hat{H}_{\mathrm{D}}^{\mathrm{np}}$, $\hat{H}_{\mathrm{A}}^{\mathrm{np}}$ and $\hat{V}_{\mathrm{DA}}^{\mathrm{np}}$ are given in terms of sums of purely nuclear and purely photonic terms:[1]

$$\hat{H}_{\mathrm{D}}^{\mathrm{np}} = \hat{H}_{\mathrm{D}}^{\mathrm{n}} + \hat{H}^{\mathrm{p}} \quad (2a)$$

$$\hat{H}_{\mathrm{A}}^{\mathrm{np}} = \hat{H}_{\mathrm{A}}^{\mathrm{n}} + \hat{H}^{\mathrm{p}} \quad (2b)$$

$$\hat{V}_{\mathrm{DA}}^{\mathrm{np}} = \hat{V}_{\mathrm{DA}}^{\mathrm{n}} + \hat{V}_{\mathrm{DA}}^{\mathrm{p}}. \quad (2c)$$

Here, the n and p superscripts identify contributions from the nuclear and photonic DOF, respectively. It should be noted that, while the nuclear Hamiltonians $\hat{H}^{\rm n}_{\rm D}$ and $\hat{H}^{\rm n}_{\rm A}$ are electronic-state specific, the corresponding photonic Hamiltonian, $\hat{H}^{\rm p}$, is not. Going forward, we will also assume that the molecular electronic coupling term satisfies the Condon approximation, such that $\hat{V}^{\rm n}_{\rm DA} = V^{\rm n}_{\rm DA}$ (a constant, as opposed to a nuclear operator). Finally, we note that the Hamiltonian of the molecular system in the cavity-free case is given by

$$\hat{H}^{\rm m} = \hat{H}^{\rm n}_{\rm D}|{\rm D}\rangle\langle{\rm D}| + \hat{H}^{\rm n}_{\rm A}|{\rm A}\rangle\langle{\rm A}| + \hat{V}^{\rm n}_{\rm DA}[|{\rm D}\rangle\langle{\rm A}| + |{\rm A}\rangle\langle{\rm D}|], \quad (3)$$

where the superscript m will be used throughout to indicate purely molecular (i.e. cavity-free) operators.

In this work, the photonic Hamiltonian and electronic coupling term are assumed to account for contributions from multiple cavity modes:[1]

$$\hat{H}^{\rm p} = \sum_{\alpha} \hat{H}^{{\rm p}\alpha} = \frac{1}{2}\sum_{\alpha}\left(\hat{p}^2_{{\rm p}\alpha} + \omega^2_{{\rm p}\alpha}\hat{q}^2_{{\rm p}\alpha}\right), \quad (4)$$

$$\hat{V}^{\rm p}_{\rm DA} = \sum_{\alpha} \hat{V}^{{\rm p}\alpha}_{\rm DA} = \sum_{\alpha} \sqrt{2\hbar\omega_{{\rm p}\alpha}}g_{{\rm p}\alpha}\,\hat{q}_{{\rm p}\alpha}. \quad (5)$$

Here, $\hat{H}^{{\rm p}\alpha}$ and $\hat{V}^{{\rm p}\alpha}_{\rm DA}$ are the contributions of the α-th cavity mode to the photonic Hamiltonian and electronic coupling term, respectively ($\alpha = 1, 2, 3, \ldots$). This should be contrasted with Ref. 1 where only a single photonic mode was assumed (i.e. the above sums over the index α reduce to a single term). $\{\hat{q}_{{\rm p}\alpha}\}$, $\{\hat{p}_{{\rm p}\alpha}\}$ and $\{\omega_{{\rm p}\alpha}\}$ are the positions, momenta and angular frequencies associated with the cavity modes and

$$g_{{\rm p}\alpha} = \sqrt{\frac{\mu^2_{\rm DA}\omega_{{\rm p}\alpha}}{2\hbar\varepsilon_0 V}}, \quad (6)$$

where $\mu_{\rm DA}$ is the donor-acceptor transition dipole moment, ε_0 is the vacuum permittivity and V is the volume of the cavity.

In the next step, we specialize to the case of a two-dimensional cavity with the z and x-directions parallel and perpendicular to the mirrors, respectively. The frequencies of the photonic modes in such a case are given by:

$$\omega_p = \frac{c}{n_r}\sqrt{k_x^2 + k_z^2}, \quad (7)$$

where k_x and k_z are wave vectors that correspond to the respective directions and n_r is the refractive index. In the x-direction we restrict ourselves to the fundamental mode $k_{0,x} = \pi/L_x$, where L_x is the width of the cavity along the x axis, while in the z-direction the modes are assumed to satisfy periodic boundary condition, such that $k_{\alpha z} = 2\alpha\pi/L_z$, where L_z is the length of the cavity along the z axis. Substituting $k_x = k_{0,x} = \pi/L_x$ and $k_{\alpha z} = 2\alpha\pi/L_z$ back into Eq. (7) yields:

$$\omega_{{\rm p}\alpha} = \sqrt{\omega^2_{\rm p0} + \alpha^2\delta_z^2}, \quad (8)$$

where $\omega_{\rm p0} = c\pi/(n_r L_x)$ is the fundamental cavity frequency along the x axis, and $\delta_z = 2c\pi/(n_r L_z)$ is assumed to be a parameter independent of L_x or $\omega_{\rm p0}$.[2,3] We also assume that $g_{{\rm p}\alpha}$ is proportional to $\sqrt{\omega_{\rm p0}\omega_{{\rm p}\alpha}}$, such that the dimensionless parameter χ defined by Eq. (9), which measures strength of the cavity-induced electronic coupling relative to that of the cavity-free electronic coupling, is independent of α:

$$\frac{1}{\chi} \equiv \frac{\hbar^2 g^2_{{\rm p}\alpha}}{(V^n_{\rm DA}\beta\hbar)^2\omega_{\rm p0}\omega_{{\rm p}\alpha}}. \quad (9)$$

III. CAVITY-MODIFIED EQUILIBRIUM FGR RATE CONSTANTS

The main assumption underlying the derivation of the cavity-modified equilibrium FGR rate constant is that the electronic coupling term, $\hat{V}^{\rm np}_{\rm DA}[|{\rm D}\rangle\langle{\rm A}| + |{\rm A}\rangle\langle{\rm D}|]$, can be treated as a small perturbation within the framework of second-order perturbation theory. This assumption, together with the assumption that the onset of rate kinetics happens on a time scale which is much faster than the time scale of the actual electronic transition (set by the inverse of the rate constant), results in the following expression for the *cavity-modified donor-to-acceptor transition rate constant*:[1,4–6]

$$k_{\rm D\to A} = \frac{1}{\hbar^2}\int_{-\infty}^{\infty} {\rm d}t\, C_{\rm D\to A}(t). \quad (10)$$

Here, $C_{\rm D\to A}(t)$ is the *cavity-modified donor-to-acceptor FGR time correlation function*, which is explicitly given by

$$C_{\rm D\to A}(t) = {\rm Tr_n Tr_p}\left[\hat{\rho}^{\rm eq}_{\rm D} e^{i\hat{H}^{\rm np}_{\rm D}t/\hbar}\hat{V}^{\rm np}_{\rm DA}e^{-i\hat{H}^{\rm np}_{\rm A}t/\hbar}\hat{V}^{\rm np}_{\rm DA}\right]. \quad (11)$$

The corresponding *cavity-free donor-to-acceptor transition rate constant* is given by

$$k^{\rm m}_{\rm D\to A} = \frac{1}{\hbar^2}\int_{-\infty}^{\infty} {\rm d}t\, C^{\rm m}_{\rm D\to A}(t), \quad (12)$$

where $C^{\rm m}_{\rm D\to A}(t)$ is the *cavity-free donor-to-acceptor FGR time correlation function*:

$$C^{\rm m}_{\rm D\to A}(t) = {\rm Tr_n}\left[\hat{\rho}^{\rm eq}_{\rm D,n} e^{i\hat{H}^{\rm n}_{\rm D}t/\hbar}\hat{V}^{\rm n}_{\rm DA}e^{-i\hat{H}^{\rm n}_{\rm A}t/\hbar}\hat{V}^{\rm n}_{\rm DA}\right]. \quad (13)$$

Here, $\rm Tr_n$ and $\rm Tr_p$ are the traces over the nuclear and photonic Hilbert spaces, respectively; $\hat{\rho}^{\rm eq}_{\rm D} = e^{-\beta\hat{H}^{\rm np}_{\rm D}}/{\rm Tr_n Tr_p}\left[e^{\beta\hat{H}^{\rm np}_{\rm D}}\right]$ is the density operator that describes the initial state of the photonic + nuclear DOF in the *cavity-confined* system, assumed to be in the thermal equilibrium state that corresponds to the donor state; $\hat{\rho}^{\rm eq}_{\rm D,n} = e^{-\beta\hat{H}^{\rm n}_{\rm D}}/{\rm Tr_n}\left[e^{\beta\hat{H}^{\rm n}_{\rm D}}\right]$ is the density operator that describes the initial state of the nuclear DOF in the *cavity-free* system, assumed to be in the thermal equilibrium state that corresponds to the donor state.[1]

The main quantity of interest in this communication is the ratio of the cavity-modified to cavity-free donor-to-acceptor transition rate constants, $\frac{k_{\rm D\to A}}{k^{\rm m}_{\rm D\to A}}$, which captures the sought after cavity-induced

relative change in the FGR rate constant. Thus, $\frac{k_{D\to A}}{k^{m}_{D\to A}} = 1$ implies no change, $\frac{k_{D\to A}}{k^{m}_{D\to A}} > 1$ implies a cavity-induced rate enhancement and $\frac{k_{D\to A}}{k^{m}_{D\to A}} < 1$ implies cavity-induced rate suppression.

Following a similar procedure to that applied to a single-mode model in Ref. 1, and taking advantage of the fact that the modes in the multiple-mode case are uncoupled, then leads to the following expression for $\frac{k_{D\to A}}{k^{m}_{D\to A}}$:

$$\frac{k_{D\to A}}{k^{m}_{D\to A}} = 1 + \sum_{\alpha} \frac{\hbar^2 g^2_{p\alpha}}{(V^{m}_{DA})^2} \langle \hat{N}(\beta\hbar\omega_{p\alpha})\rangle_{eq} \left[\frac{k^{m}_{D\to A}(\omega_{p\alpha})}{k^{m}_{D\to A}(0)} + \frac{k^{m}_{A\to D}(\omega_{p\alpha})}{k^{m}_{A\to D}(0)} \right]. \tag{14}$$

Here, $\langle \hat{N}(\beta\hbar\omega_{p\alpha})\rangle$ is the expectation value of the number of photons in the α-th cavity mode at thermal equilibrium,

$$\langle \hat{N}(\beta\hbar\omega_{p\alpha})\rangle_{eq} = \frac{1}{e^{\beta\hbar\omega_{p\alpha}} - 1}, \tag{15}$$

$k^{m}_{D\to A}(\omega_{p\alpha})$ is given by

$$k^{m}_{D\to A}(\omega_{p\alpha}) = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} dt e^{i\omega_{p\alpha} t} C^{m}_{D\to A}(t), \tag{16}$$

and $k^{m}_{A\to D}(\omega_{p\alpha})$ can be obtained from $k^{m}_{D\to A}(\omega_{p\alpha})$ by switching the labels A and D in Eqs. (12), (13), and (16). It should be noted that $k^{m}_{D\to A}(0) \equiv k^{m}_{D\to A}$ and $k^{m}_{A\to D}(0) \equiv k^{m}_{A\to D}$ are the cavity-free donor-to-acceptor and acceptor-to-donor transition rate constants, respectively. It should be noted that $\frac{k_{D\to A}}{k^{m}_{D\to A}} \to \infty$ when $V_{DA} \to 0$, which reflects the fact that the cavity-free $k^{m}_{D\to A} \to 0$ when $V_{DA} \to 0$, while the cavity-modified $k_{D\to A}$ remains finite.

Equation (14) is the main result of this communication. It represents a generalization of a similar expression obtained for the single-mode case in Ref. 1. The main difference is the sum over multiple modes on the R.H.S. of Eq. (14), which reduces to a single term in the single cavity mode case studied in Ref. 1. Since every term in the sum on the R.H.S. of Eq. (14) is non-negative, we conclude that $\frac{k_{D\to A}}{k^{m}_{D\to A}} \geq 1$, with equality in the cavity-free case. Thus, within our model, coupling to cavity modes would always enhance the rate constant, with the overall enhancement consisting of the *sum* of enhancements associated with coupling to individual cavity modes. In this communication, we will focus on the effect on the cavity-induced enhancement of the donor-to-acceptor rate constant caused by going from a single mode to multiple modes. To this end, we focus on electronic energy and charge transfer reactions whose cavity-free kinetics can be described by Marcus theory, which corresponds to a widely used approximate version of FGR-based rate theory.

IV. CAVITY-MODIFIED MARCUS THEORY RATE CONSTANTS

As is well known, the cavity-free donor-to-acceptor FGR rate constant, Eqs. (12) and (13), reduces to the Marcus theory rate constant in the short-time and high-temperature limits, provided that the nuclear dynamics satisfy Gaussian statistics.[5,6] Within this commonly assumed limit of FGR rate theory, the various cavity-free rate constants that show up on the R.H.S. of Eq. (14) are given by the following expressions:[1]

$$k^{m}_{D\to A}(0) = \frac{(V^{m}_{DA})^2}{\hbar} \sqrt{\frac{\pi}{k_B T E_r}} e^{-\frac{(\Delta E + E_r)^2}{4k_B T E_r}}, \tag{17a}$$

$$k^{m}_{A\to D}(0) = \frac{(V^{m}_{DA})^2}{\hbar} \sqrt{\frac{\pi}{k_B T E_r}} e^{-\frac{(-\Delta E + E_r)^2}{4k_B T E_r}}, \tag{17b}$$

$$k^{m}_{D\to A}(\omega_{p\alpha}) = \frac{(V^{m}_{DA})^2}{\hbar} \sqrt{\frac{\pi}{k_B T E_r}} e^{-\frac{(\Delta E - \hbar\omega_{p\alpha} + E_r)^2}{4k_B T E_r}}, \tag{17c}$$

$$k^{m}_{A\to D}(\omega_{p\alpha}) = \frac{(V^{m}_{DA})^2}{\hbar} \sqrt{\frac{\pi}{k_B T E_r}} e^{-\frac{(-\Delta E - \hbar\omega_{p\alpha} + E_r)^2}{4k_B T E_r}}. \tag{17d}$$

Here, ΔE is the *cavity-free donor-to-acceptor reaction free energy* and E_r is the *cavity-free reorganization energy*. In what follows, we will assume that the cavity-free *donor-to-acceptor* (D → A), transition is thermodynamically favorable (i.e. $\Delta E < 0$).

Following a procedure similar to that followed in Ref. 1, we define the following *multi-mode cavity enhancement function of the Marcus theory rate constant*:

$$\begin{aligned} F(\beta\hbar\omega_{p0,\beta\hbar\delta_z}) &= \chi\left(\frac{k_{D\to A}}{k^{m}_{D\to A}} - 1\right) \\ &= \sum_{\alpha} \beta^2\hbar^2\omega_{p0}\omega_{p\alpha} \langle \hat{N}(\beta\hbar\omega_{p\alpha})\rangle_{eq} \\ &\quad \times \left[\frac{k^{m}_{D\to A}(\omega_{p\alpha})}{k^{m}_{D\to A}(0)} + \frac{k^{m}_{A\to D}(\omega_{p\alpha})}{k^{m}_{A\to D}(0)} \right]. \end{aligned} \tag{18}$$

We note that the frequencies of the multiple cavity modes, $\{\omega_{p\alpha} | \alpha = 1, 2, \ldots\}$, depend on two parameters, which are determined by the experimental setup, namely ω_{p0} and δ_z [see Eq. (8)]. The results presented below are based on assuming that $\delta_z = 0.5$ a.u., which corresponds to $L_z \sim 0.1$ μm [using $\delta_z = 2c\pi/(n_r L_z)$ and assuming $n_r = 1$]. We also consider a range of ω_{p0} values such that $\beta\hbar\omega_{p0} \in [1, 50]$, which corresponds to $L_x \in [0.5 - 20]$ μm [using $\omega_{p0} = c\pi/(n_r L_x)$ and assuming $n_r = 1$ and $T = 300$ K]. The fact that the length scale of the cavity (as measrued by L_x and L_y) is in the μm range implies that they correspond to experimentally accessible microcavities. We also consider values of $\beta|\Delta E|$ and βE_r between 5 and 50. This corresponds to $|\Delta E|$ and E_r in the (0.13–1.30) eV range at room temperature, which is typical for charge transfer reactions in molecular systems.[1] In what follows, we will focus on analyzing the effect of going from a single cavity mode to multiple cavity modes on the cavity-induced enhancement of the Marcus theory rate constant.

The cavity enhancement function in Eq. (18), $F(\beta\hbar\omega_{p0}, \beta\hbar\delta_z)$, is shown in Fig. 1, as a function ω_{p0}, for different numbers of cavity modes (1, 2, 10, 100). Differet panels in this figure correspond to different values of $\beta\Delta E$ and βE_r. Also shown in Fig. 1 is the contribution from the dominant cavity mode, which corresponds to the cavity mode with the largest contribution to the sum on the R.H.S. of Eq. (18). Inspection of Fig. 1 reveals that going from a single cavity mode to multiple cavity modes can enhance the cavity-modified Marcus theory rate constant by at least an order of magnitude. Furthermore, the additional enhancement is cumulative, i.e. the enhancement results from summing over multiple terms on the R.H.S. of Eq. (18) and cannot be explained by a single dominant mode. We also note that while the dominant term corresponds to

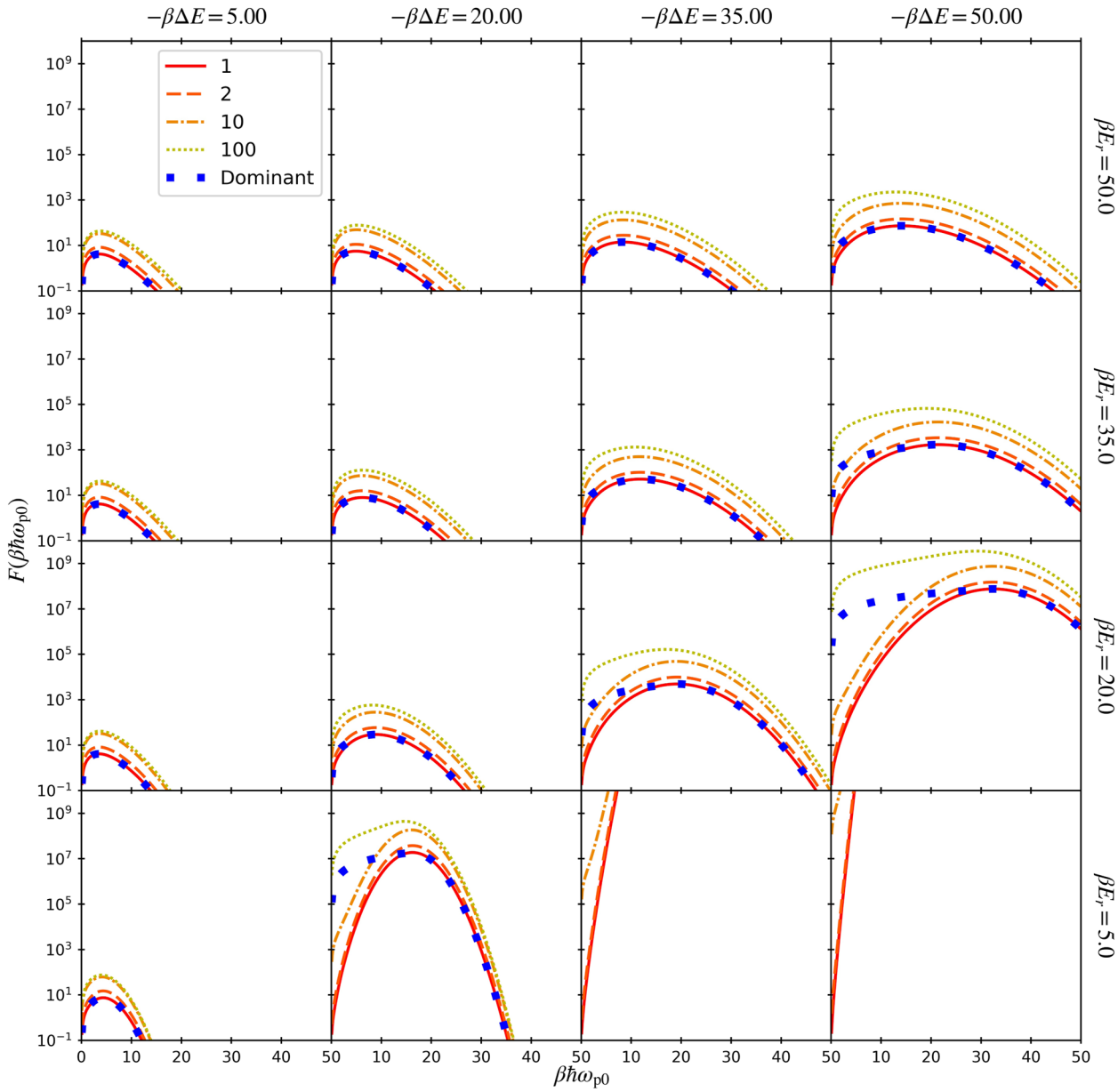

FIG. 1. Plots of $F(\beta\hbar\omega_{p0,\beta\hbar\delta_z})$ as a function of $F(\beta\hbar\omega_{p0}$, for different numbers of cavity modes (1, 2, 10, 100). Differet panels in this figure correspond to different values of $\beta\Delta E$ and βE_r. Also shown is the contribution from the dominant cavity mode, which corresponds to the cavity mode with the largest contribution to the sum on the R.H.S. of Eq. (18) (dotted blue line).

the principal cavity mode ($\alpha = 0$) at small values of $|\Delta E|$, higher frequency cavity modes become dominant at larger values of $|\Delta E|$, and even more so at larger values of E_r.

While the number of modes included in the sum on the R.H.S. of Eq. (18) is in principle infinite, a closer inspection reveals that there is an effective upper limit beyond which additional modes do not contribute significantly to sum. This is demonstrated in Fig. 2(a) which shows the ratio between the multiple-mode and single-mode enhancement functions (F_N and F_1, respectively) as a function of the number of cavity modes, N, for three different values of the principle cavity frequency, ω_{p0} (see insert), in the case where $\beta|\Delta E| = 35$ and $\beta E_r = 20$. The value of F_N/F_1 is seen to plateau beyond a certain finite number of modes ($N \sim 40$–60 in this example), which implies an effective upper cutoff frequency beyond which

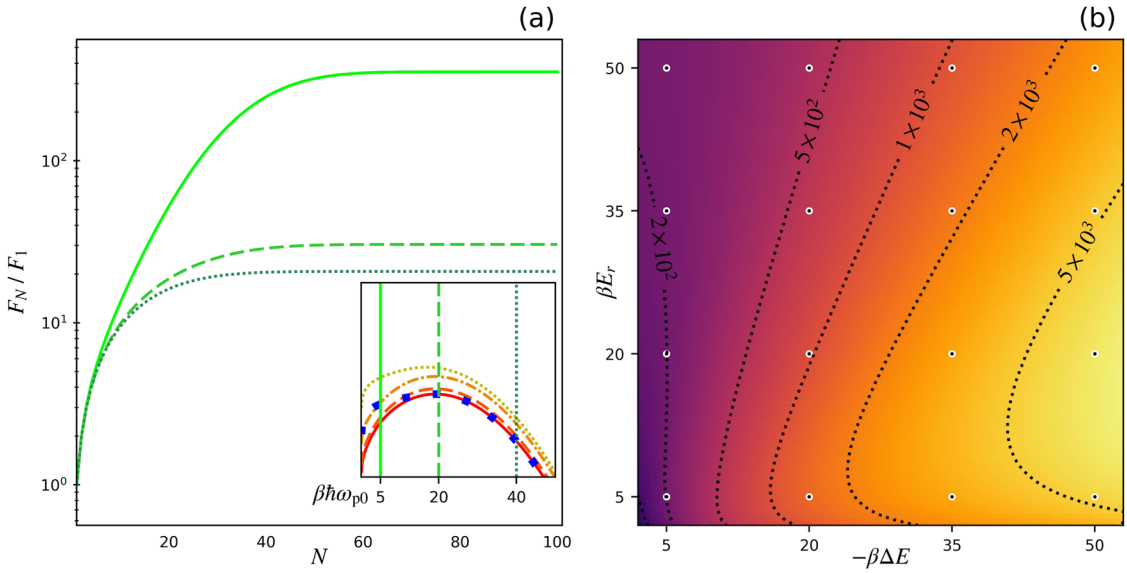

FIG. 2. (a) The ratio between the multiple-mode and single-mode enhancement functions (F_N and F_1, respectively) as a function of the number of cavity modes, N, for three different values of the principle cavity frequency, ω_{p0} (see insert), in the case where $\beta|\Delta E| = 35$ and $\beta E_r = 20$. (b) A plot of the ratio between the maximum enhancement in the case of $N = 100$ cavity modes, $F_{100}^{\max}$, and the maximum enhancement in the case of $N = 1$ (a single cavity mode), $F_1^{\max}$, as a function of βE_r and $\beta\Delta E$. The 16 black points correspond to the 16 panels of Fig. 1.

multi-mode enhancement becomes ineffective. It should also be noted that the maximum multi-mode enhancement is sensitive to the value of ω_{p0}.

Further insight can be obtained from Fig. 2(b) where we plot the ratio between the maximum enhancement in the case of $N = 100$ cavity modes, $F_{100}^{\max}$, and the maximum enhancement in the case of $N = 1$ (a single cavity mode), $F_1^{\max}$, as a function of βE_r and $\beta\Delta E$. The value of $F_{100}^{\max}/F_1^{\max}$ is seen to range between 2×10^2–5×10^3, and to increase with increasing $|\Delta E|$ and decreasing E_r. It should also be noted that the dependence on $|\Delta E|$ is significantly stronger than the dependence on E_r. We note that a similar trend was previously reported in the context of the dependence of $F_1^{\max}$ on ΔE and E_r.[1]

V. CONCLUSIONS

FGR rate theory has proven to be extremely useful for calculating electronic energy and charge transfer rates in molecular systems. The considerable recent interest in the ability to modify rates of chemical processes by placing the molecular system inside a photonic cavity therefore calls for a cavity-modified FGR rate theory. In this communication we examined the effect of going from a model that accounts for a single cavity mode in a one-dimensional cavity to a more realistic model that accounts for multiple cavity modes in a two-dimensional cavity. To this end, we extended our recently proposed theoretical framework for estimating cavity-modified equilibrium FGR rate constants[1] beyond the single cavity mode case to cases where the molecular system is coupled to multiple cavity modes.

Similarly to the single-mode framework in Ref. 1, the FGR rate constant can be calculated from cavity-free inputs in the multi-mode case. Our major finding is that accounting for multiple cavity modes can enhance the Marcus theory rate constant by several orders of magnitude in comparison to the single-mode case. Furthermore, the additional enhancement is cumulative and cannot be explained by a single dominant mode. Another finding is that the rate is affected by a finite number of cavity modes whose frequency lies below a cutoff frequency. A detailed analysis of the conditions necessary for maximizing this effect and its dependence on the reaction energy and reorganization energy was presented in the case of the Marcus limit of FGR rate theory.

While being rather general, the proposed framework is still based on a number of assumptions, including restricting ourselves to two electronic states, assuming zero permanent dipole moments and neglecting nonequilibrium effects. Extending the framework beyond those restrictive assumptions would be desirable towards closing the gap between theory and experiment in this emerging domain of chemical reactivity.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Maximlian A. C. Saller: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Software (equal); Validation

(equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Yifan Lai**: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Software (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Eitan Geva**: Conceptualization (equal); Formal analysis (equal); Funding acquisition (lead); Project administration (lead); Supervision (lead); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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