Interaction effect on adiabatic pump of charge and spin in quantum dot

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We investigate the pumped charge and spin at zero-bias by adiabatic modulation of two control parameters using the generalized quantum master equation approach. First we study all higher order effects of the pumping frequency in general systems and show the equivalence between our approach and the real-time diagrammatic approach. Next we investigate the adiabatic pump in quantum dots weakly coupled to two leads. The pumped charge (spin) is given by a summation of a time integral of the steady charge (spin) current and a geometric surface integral of the Berry-Sinitsyn-Nemenman curvature. We show that the former is generally principal if the thermodynamic parameters are modulated although it is zero if the thermodynamic parameters are fixed to zero-bias. Collinear magnetic fields with different tunable amplitudes are applied to the quantum dots and the leads. For interacting one level system, we calculate analytically the pumped charge and spin by modulating the magnetic fields and the coupling strengths to the leads in the weak and strong interacting limit. We show that the difference between these two limits appears through the factor denoting the average number of up or down spin electrons in the quantum dot.

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I. INTRODUCTION

In a mesoscopic system, even at zero-bias, a charge or spin current is induced by a slow modulation of two or more control parameters. This phenomenon, called quantum adiabatic pump, is theoretically interesting because its origins are quantum effects and non-equilibrium effects. The quantum adiabatic pump is also expected to be applied to the single electron transfer devices and the current standard.

The pumped current is described by a geometric expression in the control parameter space. In non-interacting systems, the quantum adiabatic pump had extensively been studied by the Brouwer formula, which describes the pumped charge by the scattering matrix. On the other hand it is difficult to calculate the scattering matrix in the interacting systems. In the interacting case, the Brouwer formula had only been applied in mean field treatments or in the Toulouse limit. Sinitsyn and Nemenman had studied the adiabatically pumped charge using the full counting statistics and had shown that it is characterized by a Berry-phase-like vector (Berry-Sinitsyn-Nemenman (BSN) vector). The BSN vector was applied to the spin boson system and was made connection to the excess entropy production.

Recently, the quantum pump in interacting systems have been actively researched. There are two theoretical approaches. First is the real-time diagrammatic approach which uses similar equation to the quantum master equation (QME). Particularly Ref. had derived a geometric expression similar to the Brouwer formula and the BSN vector. The second is the generalized quantum master equation approach proposed in Ref. which gives the BSN vector from the GQME with the Markov approximation. The GQME approach has several advantages to the Brouwer formula:

(i) It can treat the Coulomb interaction easily. (ii) The finite temperature effects and the bias effects are also treatable. (iii) We can calculate the higher order cumulants of the pumped charge from the cumulant generating function. The derived formula of pumped charge depends on the approximation used for the QME. The Born-Markov approximation with or without the rotating wave approximation (RWA) are frequently used. The QME in Born-Markov approximation without RWA sometimes violates the non-negativity of the system reduced density operator. The QME of the RWA or the Coarse-graining approximation (CGA) is the Lindblad type which guarantees the non-negativity. Yuge et al. studied the pumped charge coming from the BSN curvatures by adiabatic modulation of the thermodynamic parameters (the chemical potentials and the temperatures) in spinless quantum dots weakly coupled to two spinless leads and showed that the BSN curvatures are zero in non-interacting system although they are nonzero for finite interaction.

In this paper, we first generalize the GQME approach to multi-counting fields to calculate spin current. We then study the non-adiabatic effects in general systems and relations between the GQME approach and the real-time diagrammatic approach in the adiabatic condition. Next we explain the model to be considered. We consider quantum dots strongly coupled to two leads. We apply collinear magnetic fields with different amplitudes to the quantum dots and the leads. The dynamic parameters (the strengths of these magnetic fields and the coupling strengths to the leads) are control parameters. We use the RWA defined as a long coarse-graining time limit of the CGA to the GQME. In §IV and §V we consider non-interacting and interacting systems respectively. First, we show (in §V.B) the
time integral of a steady current is generally principal if the thermodynamic parameters (the chemical potentials and the temperatures of leads) are modulated (as considered in Refs. 25, 28, 32, 33). The condition that the contribution of the steady currents vanish had not been previously clarified. If we want to detect or apply the BSN curvatures easily, we should fix the thermodynamic parameters to zero-bias. Next in a one level system with the Coulomb interaction $U$, we analytically calculate the BSN curvatures of spin and charge induced by the dynamic parameters in the weak $|\nabla V C|$ and strong $|\nabla V C|$ interacting limit ($U \to 0, \infty$). The difference between the results for $U = 0$ and $U = \infty$ appears through the factor denoting the average number of up or down spin electrons of the quantum dot. In $|\nabla V C|$ we show and discuss the contour plots of BSN curvatures evaluated numerically. At last, we summarize this paper with discussions (§VI).

### II. GQME

In this section, we consider general systems weakly coupled to non-interacting (fermionic or bosonic) baths. The model we use to a concrete calculation is explained at §II.

In §IIA we explain the GQME method using the Liouville space $21,39$ (Appendix A). This method is a generalization to multi-counting fields of Ref. 32. In §IIIB we study non-adiabatic effect, and show the equivalence to the method of Ref. 29.

#### A. Derivation of GQME

Consider a cyclic modulation of the control parameters with a period $\tau$. At $t = 0$ and $t = \tau$, we perform projection measurements of $\mu$-th time-independent observables $\{\hat{O}_\mu\}$ of baths which commute with each other. $\Delta \hat{O}_\mu = \hat{O}_\mu(\tau) - \hat{O}_\mu(0)$ denotes the difference of the outcomes $\{\hat{O}_\mu(\tau)\}$ at $t = \tau$ and the outcomes $\{\hat{O}_\mu(0)\}$ at $t = 0$. The Fourier transform of the joint probability density distribution $P_\tau(\{\Delta \hat{O}_\mu\})$, $Z_\tau(\{\chi_\mu\}) = \int \prod \mu d\Delta \hat{O}_\mu P_\tau(\{\Delta \hat{O}_\mu\}) e^{\sum \mu \chi_\mu \Delta \hat{O}_\mu}$, is the generating function. Here, $\chi_\mu$ are fields for $\hat{O}_\mu$. $Z_\tau(\{\chi_\mu\})$ is given by $Z_\tau(\{\chi_\mu\}) = \text{Tr}_{\text{tot}}[\rho^{\chi}(t = \tau)]$ using an operator of total systems $\rho^{\chi}(t = \tau)$. Here, $\chi$ denotes the set of the counting fields $\{\chi_\mu\}$. In Appendix B we derive the generalized quantum master equation (GQME) (i.e., the equation of motion of $\rho^{\chi}(t) = \text{Tr}_{S}[\rho^{\chi}(t)]$ where $\text{Tr}_{S}$ denotes a trace over bath degree of freedom) from the equation of motion of $\rho^{\chi}(t)$, $\rho^{\chi}(t)$ provides the generating function $Z_\tau(\{\chi_\mu\}) = \text{Tr}_{S}[\rho^{\chi}(t = \tau)]$. The GQME is

$$\frac{d\rho^{\chi}(t)}{dt} = \dot{K}^{\chi}(\alpha_t)\rho^{\chi}(t),$$

(1)

where $\dot{K}^{\chi}(\alpha_t)$ is the Liouvillian modified by $\chi$, and $\alpha_t$ is the value of the set of the control parameters at time $t$. At $\chi = 0$, the GQME becomes the quantum master equation (QME)

$$\frac{d\rho(t)}{dt} = \dot{K}(\alpha_t)\rho(t),$$

(2)

where $\rho(t) = \text{Tr}_{B}[\rho_{\text{tot}}(t)]$ is the system reduced density operator and $\rho_{\text{tot}}(t)$ is the total system state. $\dot{K}(\alpha_t)$ equals to $\dot{K}^{\chi}(\alpha_t)$ at $\chi = 0$. In the following, a symbol $X$ without $\chi$ denotes $X^\chi|_{\chi=0}$. The initial condition of the GQME is $\rho^{\chi}(0) = \rho(0) = \text{Tr}_{B}[\rho_{\text{tot}}(0)]$. In particular, we suppose $\rho_{\text{tot}}(0) = \rho(0) \otimes \rho_B(\alpha_0)$ with the grand canonical or canonical distribution of the leads, $\rho_B(\alpha_0)$.

In the Liouville space (Appendix A), the left and right eigenvalue equations of the Liouvillian are

$$\dot{K}^{\chi}(\alpha)|\rho^{\chi}(\alpha)>= = \lambda_{\alpha}^{\chi}|\rho^{\chi}(\alpha)>,$$

$$\langle|\rho^{\chi}(\alpha)|\dot{K}^{\chi}(\alpha)\rangle = \lambda_{\alpha}^{\chi}\langle|\rho^{\chi}(\alpha)>,$$

(3)

(4)

The left eigenvectors $l_{\alpha}^{\chi}(\alpha)$ and the right eigenvectors $\rho_{\alpha}^{\chi}(\alpha)$ (in fact these are operators) satisfy $\langle|\rho^{\chi}(\alpha)|\rho^{\chi}(\alpha)> = \delta_{\alpha}$. Here, $\alpha$ denotes arbitrary values of the set of the control parameters. We assign the label $n = 0$ for the eigenvalue with the maximum real part. At limit of $\chi \to 0$, $\lambda_{\alpha}^{\chi}(\alpha)$ becomes 0 and $\langle|\rho^{\chi}(\alpha)>$ becomes $\langle|1\rangle$, i.e., $l_{0}(\alpha) = 1$. The conservation of the probability $\frac{d}{dt}\langle|1\rangle\rangle = \langle|1|\dot{K}(\alpha_t)|\rho(t)> = 0$ leads $\langle|1\dot{K}(\alpha) = 0$. In addition, $n = 0$ mode right eigenvector $|\rho_0(\alpha)\rangle$ determined by $\dot{K}(\alpha)|\rho_0(\alpha)> = 0$ represents the steady state; if the control parameters are fixed to $\alpha$, the state $\rho(t)$ converges to $\rho_0(\alpha)$ at $t \to \infty$. In general, the solution of the GQME Eq. (1) is expanded as

$$|\rho^{\chi}(t)> = \sum c_{n}^{\chi}(t)e^{\Lambda_{n}^{\chi}(t)}|\rho_{n}^{\chi}(\alpha_t)>$$

(5)

where $\Lambda_{n}^{\chi}(t) = \int_{0}^{t} ds \lambda_{n}^{\chi}(\alpha_s)$. The coefficients $c_{n}^{\chi}(t)$ obey

$$\frac{d}{dt} c_{n}^{\chi}(t) = -\langle|l_{m}^{\chi}(\alpha_t)|\frac{d}{dt} |\rho_{m}^{\chi}(\alpha_t)>\rangle c_{n}^{\chi}(t) + \sum_{m \neq n} c_{m}^{\chi}(t) \chi e^{\Lambda_{m}^{\chi}(t)-\Lambda_{n}^{\chi}(t)}\frac{\langle|l_{n}^{\chi}(\alpha_t)|\dot{K}^{\chi}(\alpha)\langle|l_{m}^{\chi}(\alpha_t)|\rho_{n}^{\chi}(\alpha_t)>\rangle}{\lambda_{n}^{\chi}(\alpha_t)-\lambda_{m}^{\chi}(\alpha_t)}.$$

(6)

In this section, we consider sufficiently slow modulation of the control parameters. Hence, the changes of $c_{n}^{\chi}(t)$ are also very slow. The effects of fast modulation is considered in the next section. For $m = 0$, the second term of the right side of Eq. (6) exponentially damps as a function of time. The relaxation time of the system ($\tau_S$) is the order of $\Gamma^{-1}$ where $\Gamma$ is typical a value of
the linewidth function defined later. Assuming the cycle time $\tau$ is much larger than $\tau_S$, we obtain

$$c_0^\beta(\tau) = c_0^\beta(0) \exp \left[ - \int_0^\tau dt \left( \frac{d}{dt} \rho_0^\beta(\alpha_t) \right) \right],$$

(7)

and $c_0^\beta(\tau) e^{\lambda_0^\beta(\tau)} \approx 0$ ($n \neq 0$). Using the initial condition $\rho^\beta(0) = \rho(0)$, we obtain $c_0^\beta(0) = \langle \rho_0^\beta(\alpha_0) | \rho(0) \rangle$. Substituting these equations into Eq. (3), we obtain

$$|\rho^\beta(\tau)| \approx \langle \rho_0^\beta(\alpha_0) | \rho(0) \rangle e^{-\int_0^\tau dt \left( \frac{d}{dt} \rho_0^\beta(\alpha_t) \right)} \times e^{\lambda_0^\beta(\alpha_t) |\rho_0^\beta(\alpha_t)|},$$

(8)

and the cumulant generating function $S_\tau(\chi) = \ln Z_\tau(\chi) = \ln \langle |\rho^\beta(\tau)| \rangle$:

$$S_\tau(\chi) = \int_0^\tau dt \lambda^\beta_0(\alpha_t) - \int C da^n \langle \rho_0^\beta(\alpha_t) | \frac{\partial |\rho_0^\beta(\alpha)\rangle}{\partial a^n} \rangle + \langle \rho_0^\beta(\alpha_t) | \rho(0) \rangle + \ln \langle |\rho_0^\beta(\alpha_t)| \rangle.$$

(9)

Here, $C$ is the trajectory from $\alpha_0$ to $\alpha_\tau$, $a^n$ are the $n$-th component of the control parameters and the summation symbol $\sum_n$ is omitted. The averages $\langle \Delta \rho_0^\beta(\mu) \rangle = \frac{\partial S_\tau(\chi)}{\partial \alpha_\mu(\chi)} |_{\chi=0}$ are

$$\langle \Delta \rho_0^\beta(\mu) \rangle = \langle \rho_0^\beta(\alpha_t) | \frac{\partial |\rho_0^\beta(\alpha)\rangle}{\partial a^n} \rangle.$$ 

(10)

The third and fourth terms of the right side of Eq. (10) can be described as a surface integral over the surface $S$ enclosed by $C$ using the Stokes theorem:

$$\langle \Delta \rho_0^\beta(\mu) \rangle = \langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Steady}}^S + \langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Berry}}^S,$$

(12)

$$\langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Steady}}^S = \int_0^\tau dt \lambda_0^\beta(\alpha_t),$$

(13)

$$\langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Berry}}^S = - \int_S da^m \wedge da^n \frac{1}{2} F_{mn}^\mu(\alpha).$$

(14)

Here, $\wedge$ is the wedge product and the summation symbol $\sum_{m,n}$ is omitted. BSN curvature $F_{mn}^\mu(\alpha)$ is given by

$$F_{mn}^\mu(\alpha) = \frac{A_{mn}^\mu(\alpha)}{\partial a^m} - \frac{A_{mn}^\mu(\alpha)}{\partial a^n}.$$ 

(15)

The second term of Eq. (12) was given by Yuge et al. However, they did not consider $\langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Steady}}^S$. In IV (16) we show that this contribution is generally principal if the thermodynamic parameters are modulated although the steady currents $\lambda_0^\beta(\alpha_t)$ are zero if the thermodynamic parameters are fixed to zero-bias.

**B. Non-adiabatic effect and BSN vector**

In this section, we consider non-adiabatic effect, which had been researched recently. If the modulation of the control parameters are not adiabatic, the difference between the state and the steady state, $\rho^\beta(\tau) - \rho_0^\beta(\alpha(t)) = \sum_n \rho_n^\beta(\alpha(t))e^{\lambda_0^\beta(\tau)}\rho_n(\alpha(t))$, is important. The coefficients $c_n(\tau)$ are given by solving Eq. (6). Reference [11] had studied the adiabatic condition, where $\{c_n(t)\}_{n \neq 0}$ are negligible. In contrast, we do not treat $\{c_n(t)\}_{n \neq 0}$ explicitly, instead, we use the pseudo-inverse of the Liouvillian.

The formal solution of the GQME Eq. (1) is

$$|\rho^\beta(\tau)| = T \exp \left[ \int_0^\tau ds \hat{K}^\beta(\alpha_s) \right] |\rho^\beta(0)|,$$

(16)

where $T$ denotes the time-ordering operation. Using this, we obtain the averages

$$\langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Steady}}^S = \int_0^\tau dt \lambda_0^\beta(\alpha_t) - \int C da^n \langle \rho_0^\beta(\alpha_t) | \frac{\partial |\rho(0)\rangle}{\partial a^n} \rangle,$$

(17)

$$\langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Berry}}^S = - \int_S da^m \wedge da^n \frac{1}{2} F_{mn}^\mu(\alpha).$$

(18)

Thus the currents $I_\mu(t)$ are given by

$$I_\mu(t) = \langle |\rho^\beta(\tau)| \rangle_{\text{Steady}}^S + \langle \Delta \rho_0^\beta(\mu) \rangle_{\text{Berry}}^S,$$

(19)

without any approximation. Substituting this with $\rho(t) \approx \rho(0)$ into Eq. (17), we obtain Eq. (10). The third and fourth terms of Eq. (10) correspond to $- \int_0^\tau dt \langle \rho_0^\beta(\alpha_t) | \frac{d}{dt} |\rho(0)\rangle\rangle$. The currents $I_\mu(t)$ are also written by

$$I_\mu(t) = \langle |W_\mu(\alpha)\rangle |\rho(0)\rangle\rangle,$$

(20)

where $W_\mu(\alpha)$ are the current operators defined by $\langle |W_\mu(\alpha)\rangle \rangle = \langle |\rho^\beta(\tau)| \rangle_{\text{Steady}}^S$. Using Eq. (18), the steady currents are given by

$$\langle |W_\mu(\alpha)\rangle |\rho(0)\rangle\rangle = \lambda_0^\beta(\alpha) = I_{\text{Steady}}^\beta(\alpha).$$

(21)

In the quantum dots weakly coupled to two leads, the electric current operator (i.e., $W_\mu(\alpha)$ corresponding to
the electric current) coincides with Ref. [40] in Born-Markov approximations without or within RWA.

The QME Eq. (2) is equivalent to
\[
\frac{d}{dt} |\rho_0(\alpha_t)| + \frac{d}{dt} |\rho^a(t)| = \dot{K}(\alpha_t)|\rho^a(t)|,
\]
because \(\dot{K}(\alpha_t)|\rho_0(\alpha_t)| = 0\). Applying the pseudo-inverse \(\mathcal{R}(\alpha)\) defined by \(\mathcal{R}(\alpha)\dot{K}(\alpha) = 1 - |\rho_0(\alpha)|\langle1\rangle\) to this equation, we obtain
\[
|\rho^a(t)| = \mathcal{R}(\alpha) \frac{d}{dt} |\rho_0(\alpha_t)| + \mathcal{R}(\alpha) \frac{d}{dt} |\rho^a(t)|
= \sum_{n=1}^{\infty} \left[ \mathcal{R}(\alpha) \frac{d}{dt} \right]^n |\rho_0(\alpha_t)| \equiv \sum_{n=1}^{\infty} |\rho^{a(n)}(t)|,
\]
using \(\langle1|\rho^a(t)|\rangle = 0\). Submitting Eq. (22) to Eq. (20), we finally reach
\[
I_{\mu}(t) = I_{\mu}^{\text{Steady}}(\alpha_t) + \sum_{n=1}^{\infty} I_{\mu}^{a(n)}(t),
\]
where \(I_{\mu}^{a(n)}(t) = \langle1|W_{\mu}(\alpha_t)|\rho^{a(n)}(t)\rangle\).

Let’s consider the relation between Eq. (19) and Eq. (23). In [11A] we used adiabatic approximation Eq. (8), which becomes \(|\rho(t)| \approx |\rho_0(\alpha_t)|\) at \(\chi = 0\). Submitting it to Eq. (20), we obtain \(I_{\mu}(t) \approx I_{\mu}^{\text{Steady}}(t)\). So, we cannot obtain non-adiabatic currents \(\sum_{n=1}^{\infty} I_{\mu}^{a(n)}(t)\). However, from \(\chi_{\mu}\)-derivative of Eq. (8), we obtain
\[
I_{\mu}(t) \approx \chi_{\mu}(\alpha_t) - \langle0|\mu(\alpha_t)|d\rangle \frac{d}{dt} |\rho_0(\alpha_t)|.
\]
This is equivalent to Eq. (10) for \(\rho(0) = \rho_0(\alpha_0)\). Equation (24) means
\[
I_{\mu}^{a(1)}(t) = -\langle0|\mu(\alpha_t)|d\rangle \frac{d}{dt} |\rho_0(\alpha_t)|.
\]
In fact, it must be equivalent to
\[
I_{\mu}^{a(1)}(t) = \langle1|W_{\mu}(\alpha_t)\mathcal{R}(\alpha) \frac{d}{dt} |\rho_0(\alpha_t)|,
\]
hence,
\[
\langle1|W_{\mu}(\alpha)\mathcal{R}(\alpha) = -\langle0|\mu(\alpha)|+\rangle c_{\mu}(\alpha)\langle1|\mathcal{R}(\alpha)\rangle
\]
is requested. Here, \(c_{\mu}(\alpha)\) are unknown constants. Eq. (27) was shown by Sagawa, et al. [24] for a single counting field. Eq. (19) and Eq. (23) are identical because of Eq. (21) and Eq. (27).

It is important to notice that relations between the QGME approach and the real-time diagrammatic approach [25-27]. In the real-time diagrammatic approach, \(\varphi_{\chi}(t) = \langle\chi|\rho_S(t)|\chi\rangle\) are governed by the generalized master equation (GME)
\[
\frac{d}{dt} \varphi_{\chi}(t) = \sum_{\eta} \int_{-\infty}^{t} dt^\prime W_{\chi\eta}(t, t^\prime) \varphi_{\eta}(t^\prime),
\]
where \(\chi\) are the number states of the system. The kernel \(W_{\chi\eta}(t, t^\prime)\) can include the higher order contribution of the tunneling interaction between baths and the system. In the GME, \(\varphi_{\eta}(t^\prime)\) can be approximated by \(\varphi_{\eta}(t^\prime) = (p_{\eta}(t^\prime) + (t - t^\prime)^{-1}\frac{dp_{\eta}(t^\prime)}{dt})^{26}\). By the way, up to the second order of the tunneling interaction (in the following we consider this level of approximation), the charge or spin current \(I_{\mu}(t)\) is given by
\[
I_{\mu}(t) = \sum_{\chi, \eta} w_{\chi\eta}(\mu(\alpha_t)p_{\eta}(t).
\]
This equation and \(w_{\chi\eta}(\mu(\alpha_t))\) respectively correspond to our Eq. (20) and \(W_{\mu}(\alpha_t)\). If the control parameters are fixed to \(\alpha_t\), the kernel becomes \(W_{\chi\eta}(\alpha_t, t - t^\prime)\) and the steady state is \(p_{\eta}(\alpha_t)\). \(p_{\eta}(\alpha_t)\) satisfy \(\sum_{\eta} K_{\chi\eta}^{(0)}(\alpha_t)p_{\eta}^{(0)}(\alpha_t) = 0\) and \(\sum_{n} p_{\eta}^{(0)}(\alpha_t) = 1\). Here,
\[
K_{\chi\eta}^{(0)}(\alpha_t) = \int_{-\infty}^{t} dt^\prime W_{\chi\eta}(\alpha_t, t - t^\prime)\text{ is the Liouvillean corresponding to our } \dot{K}(\alpha_t).
\]
The conservation of the probability leads \(\sum_{\chi} K_{\chi\eta}^{(0)}(\alpha_t) = 0\), which corresponds to our \(\langle1|\dot{K}(\alpha_t)\rangle = 0\). Substituting \(p_{\eta}(t) = p_{\eta}^{(0)}(\alpha_t) + \sum_{n} p_{\eta}^{(n)}(\alpha_t)\) into Eq. (29), we obtain
\[
I_{\mu}(t) = \sum_{k=0}^{\infty} I_{\mu}^{(k)}(t), I_{\mu}^{(k)}(t) = \sum_{\chi, \eta} w_{\chi\eta}(\mu(\alpha_t)p_{\eta}^{(k)}(t),
\]
with \(p_{\eta}^{(0)}(\alpha_t) \equiv p_{\eta}^{(0)}(\alpha_t)\). The GME Eq. (28) is expanded by the pumping frequency and its first order part
\[
\frac{dp_{\eta}^{(0)}(t)}{dt} = \sum_{\eta} K_{\chi\eta}^{(0)}(\alpha_t)p_{\eta}^{(1)}(t),
\]
leads to
\[
p_{\eta}^{(1)}(t) = \sum_{\chi} R_{\chi\eta}(\alpha_t) \frac{dp_{\eta}^{(0)}(t)}{dt}
\]
Here, \(R_{\chi\eta}(\alpha_t)\) is the pseudo-inverse of \(K^{(0)}(\alpha_t)\) corresponding to our \(\mathcal{R}(\alpha_t)\) and it is given by
\[
R_{\chi\eta}(\alpha_t) = (\tilde{K}^{-1})_{\chi\eta}, \tilde{K}_{\chi\eta} = K^{(0)}_{\chi\eta} - K^{(0)}_{\chi\eta}.
\]
Substituting Eq. (22) into Eq. (30), we obtain
\[
I_{\mu}^{(1)}(t) = \sum_{\chi} \varphi_{\chi}^{(0)}(\alpha_t) \frac{dp_{\eta}^{(0)}(t)}{dt},
\]
where \(\varphi_{\chi}^{(0)}(\alpha_t) = \sum_{\zeta, \eta} w_{\zeta\eta}(\alpha_t) R_{\chi\eta}(\alpha_t).
\]
and Eq. [26]. In the GQME approach, $I^{(1)}_μ(t)$ is given by Eq. [26] which is identical to Eq. [26]. Because of these relations, the real-time diagrammatic approach is equivalent to the GQME approach in the calculation up to the second order of the tunneling interaction. In fact, the GQME approach can treat the higher order cumulants easily.

Although the BSN phase (i.e., the argument of the exponential function of Eq. [17]) is derived under the adiabatic condition, its origin is probably a non-adiabatic effect. Because Eq. [25] shows that the BSN phase has the information of non-adiabatic part of the QME ($\rho^\alpha(t) = \rho(t) - \rho_0(\alpha_i)$). In the expansion of $I^\mu_n(t)$ obtained from a substitution of $\delta(t) = \rho_0(\alpha_i) + \sum_{n=1}^\infty \rho^{\alpha(n)}(t) = \sum_{n=0}^\infty \rho^{\alpha(n)}(t)$ into Eq. [19], n-th ($n = 0, 1, \cdots$) order non-adiabatic solution, $\rho^{\alpha(n)}(t)$, gives $(n + 1)$-th order non-adiabatic currents $I^\mu_n(t)$ because of Eq. [27].

Thus the GQME approach picks out one higher order non-adiabatic information from the solution of the QME. The discoveries of this relation and the relationship between the GQME approach and the real-time diagrammatic approach \cite{29} are one of the most important results of this paper.

III. MODEL

We consider quantum dots (denoted by $S$) weakly coupled to two leads. The total Hamiltonian is $H_{\text{tot}}(t) = H_S(t) + \sum_{b=L,R} H_b(t) + H_{1b}(t)$. Here, $H_S(t)$ is the system (quantum dots) Hamiltonian, $H_b(t)$ is the Hamiltonian of lead $b = L, R$, and $H_{1b}(t)$ is the tunneling interaction Hamiltonian between $S$ and lead $b$. The leads and the system are applied collinear magnetic fields with different amplitudes. The leads are noninteracting:

$$H_b(t) = \sum_{k, \sigma} (\varepsilon_{bk} + \sigma g_b B_b(t)) c_{bk\sigma}^\dagger c_{bk\sigma}.$$  \hspace{1cm} (35)

Here, $\sigma = \uparrow, \downarrow = \pm 1$ is spin label, $g_b = \frac{1}{2} \mu_B g_b^* b^*$ is the $g$-factor of lead $b$, $\mu_B$ is the Bohr magneton and $B_b(t)$ is the strength of magnetic field of lead $b$. $c_{bk\sigma}^\dagger (c_{bk\sigma})$ is the creation (annihilation) operator of an electron with spin $\sigma$ and momentum $k$ in lead $b$. The system Hamiltonian is

$$H_S(t) = \sum_{n, m, s, s'} \varepsilon_{ns, ms'}(B_S(t)) a_{ns}^\dagger a_{ms'} + H_C,$$  \hspace{1cm} (36)

where $a_{ns}^\dagger$ is the creation operator of an electron with orbital $n$ and spin $s$. $\varepsilon_{ns, ms'}(B_S(t))$ means the energy of the election for $n = m, s = s'$. The tunneling amplitudes between orbitals for $(n, s) \neq (m, s')$ which depend on the magnetic field of the system. $H_C$ denotes Coulomb interaction. The tunneling interaction Hamiltonian is

$$H_{1b}(t) = \sum_{k, \sigma, n, s} \sqrt{\Delta_b(t)} v_{bk\sigma, ns} a_{ns}^\dagger c_{bk\sigma} + \text{h.c.},$$  \hspace{1cm} (37)

where $\Delta_b(t)$ is a dimensionless parameter, and $v_{bk\sigma, ns}$ is the tunneling amplitude.

We assume $B_S, B_{L/R}$ and $\Delta_{L/R}$ are control parameters (denoted $\alpha' = \{B_S, B_{L/R}, \Delta_{L/R}\}$) and are called the dynamic parameters. The thermodynamic parameters (the chemical potentials and inverse temperatures of leads, $\{\mu_b\}$ and $\{\beta_b\}$) are also considered as control parameters in \cite{IV B} and \cite{IV}. We denote $\alpha'' = \{\beta_b, \mu_b\} = \{L, R\}$ and $\alpha = \alpha' + \alpha''$. Yuge et al.\cite{22} chose the set of control parameters as only $\alpha''$. But we are interested in $\alpha'$ with the reason explained in \cite{IV B}.

We choose the measured observables $\{O_\mu\} = \{N_{b\sigma}\}_{\sigma = \uparrow, \downarrow}$ with $N_{b\sigma} = \sum_k c_{bk\sigma}^\dagger c_{bk\sigma}$. The pumped charge (spin) of lead $b$ are given by $(\Delta N_{b\uparrow}) + (-)(\Delta N_{b\downarrow})$. $(\Delta N_{b\sigma})$ are calculated by Eq. [12].

In the following, we apply the GQME with rotating wave approximation (RWA). In this paper, the RWA is defined as the limit of $T \rightarrow \infty$ of the coarse-graining approximation (CGA)\cite{35,36} explained in Appendix \textbf{B}. If $H_S$ is time-independent, this RWA is equivalent to usual RWA\cite{33}.

In \textbf{IV C} and \textbf{V} we consider a one level system

$$H_S(t) = \sum_s \omega_s(B_S(t)) a_s^\dagger a_s + U a_s^\dagger a_s^\dagger a_s,$$  \hspace{1cm} (38)

as a special case of Eq. [36]. Here, $\omega_s(B_S) = \omega_0 + s g_S B_S$ with $\omega_0$ the electron energy at $B_S = 0$ and $g_S = \frac{1}{2} \mu_B g_S^*$ where $g_S^*$ is the $g$-factor of the quantum dot.

IV. NON-INTERACTING SYSTEM

In this section, we consider a non-interacting system ($H_C = 0$). The system Hamiltonian Eq. [36] can be diagonalized

$$H_S = \sum_{i=1}^{2N} \omega_i b_i^\dagger b_i,$$  \hspace{1cm} (39)

by a unitary transform $a_{ns} = \sum_{i=1}^{2N} U_{ns,i} b_i$. The tunneling interaction Hamiltonian Eq. [37] is

$$H_{1b} = \sum_{k, \sigma, i} W_{bk\sigma, i} b_i^\dagger c_{bk\sigma} + \text{h.c.},$$  \hspace{1cm} (40)

with $W_{bk\sigma, i} = \sum_{s, i} \sqrt{\Delta_b(t)} v_{bk\sigma, ns} U_{ns,i}$.

In \textbf{IV A} the Liouvillian and its steady state are explained. In \textbf{IV B} we consider the contribution of Eq. [13] and show that this cannot be neglected in general if the chemical potentials and the temperatures are not fixed. In \textbf{IV C} we calculate the BSN curvatures for two combinations of modulated control parameters ($B_L, B_S$) and ($\Delta_L, B_S$).
A. Liouvillian

The Liouvillian in the RWA is given by

\[
\hat{K}^X(\alpha) = \sum_{i=1}^{2N} \hat{K}_i^X(\alpha),
\]

\[
\hat{K}^X(\alpha)_\bullet = -i [\omega b_i^\dagger b_i, \bullet] + \hat{\Pi}_i^X(\alpha)_\bullet + \hat{\Pi}^* i(\alpha)_\bullet, \tag{42}
\]

if \{\omega_i\} are not degenerated. Here, super-operators \(\hat{\Pi}_i^X(\alpha)_\bullet\) and \(\hat{\Pi}^* i(\alpha)_\bullet\) operate to an arbitrary operator \(\bullet\) as

\[
\hat{\Pi}_i^X(\alpha)_\bullet = \sum_b \left\{ \Phi_{b,i}^+ \chi b_i^\dagger b_i^\dagger - \frac{1}{2} \Phi_{b,i}^+ \bullet b_i \Phi_{b,i}^+ b_i^\dagger + \Phi_{b,i}^- \chi b_i^\dagger b_i^\dagger - \frac{1}{2} \Phi_{b,i}^- \bullet b_i \Phi_{b,i}^- b_i^\dagger \right\}, \tag{43}
\]

\[
\hat{\Pi}^* i(\alpha)_\bullet = i [\Omega_i(\alpha) b_i^\dagger b_i, \bullet], \tag{44}
\]

with

\[
\Phi_{b,i}^{\pm,X} = 2\pi \sum_{k,\sigma} |W_{bks,i}|^2 f_b^\pm(\omega_i) e^{i\pi \chi_b \sigma} \delta(\epsilon_{bk} + \sigma g_b B_b - \omega_i),
\]

\[
\Psi_{b,i}^{\pm,X} = 2\pi \sum_{k,\sigma} |W_{bks,i}|^2 f_b^\pm(\omega_i) \rho_{bk}^\sigma + \sigma g_b B_b - \omega_i,
\]

and \(\Omega_i(\alpha) = \frac{1}{2} \sum_b \left( \Psi_{b,i}^- + \Phi_{b,i}^+ \right) \). Here, \(f_b^\pm(\omega) = [e^{i\beta_b (\omega - \mu_b)} + 1]^{-1}\) is the Fermi distribution function, \(f_b^-(\omega) = 1 - f_b^+(\omega)\), \(\chi_b\) is the counting field for \(N_{bs}\) and \(P\) denotes the Cauchy principal value. The matrix representation of \(\hat{K}_i^X(\alpha)\) (see Appendix A) by the number states of \(b_i^\dagger b_i\) \{\{0\} and \{1\}\} is a \(4 \times 4\) matrix which is block diagonalized to \{\{0\}i(0)\{1\}i(1)\}-space and \{\{0\}i(0)\{1\}i(1)\}-space. The \{\{0\}i(0)\{1\}i(1)\}-part is given by

\[
K_i^X(\alpha) = \left( \begin{array}{cc} -\Phi_{i}^+ & \Phi_{i}^- \chi \vspace{2mm} \\
-\Phi_{i}^- \chi & -\Phi_{i}^+ \end{array} \right) \begin{array}{c} \langle 0 \rangle_i \\
\langle 1 \rangle_i \end{array}, \tag{45}
\]

with \(\Phi_{i}^{\pm,X} = \sum_b \Phi_{b>i}^{\pm,X}\). The \{\{0\}i(0)\{1\}i(0)\}-part does not relate to the steady state of \(\hat{K}_i^X(\alpha)\). The eigenvalue of the steady state of \(\hat{K}_i^X(\alpha)\) is given by

\[
\lambda_{i,0}(\alpha) = -\Phi_{i}^+ + \Phi_{i}^- \frac{\chi}{2} + \sqrt{D_i^X(\alpha)}, \tag{46}
\]

with \(D_i^X(\alpha) = (\Phi_{i}^+ + \Phi_{i}^-)^2 / 4 - [\Phi_{i}^+ \Phi_{i}^- - \Phi_{i}^- \chi \Phi_{i}^+]\). The corresponding left and right eigenvectors are \(|\rho_{i,0}(\alpha)\rangle = C_{i,0}(\alpha)|0\rangle_i + E_{i,0}(\alpha)|1\rangle_i\) and \(\langle \rho_{i,0}(\alpha) | = i \langle 0 | + \nu_{i,0}(\alpha) \langle 1 |\) with \(C_{i,0}(\alpha) = \frac{\Phi_{i}^- \chi \nu_{i,0}(\alpha)}{[\lambda_{i,0}^2 + \Phi_{i}^+ \Phi_{i}^- \chi^2 + \Phi_{i}^+ \Phi_{i}^- \chi^2]}\) and \(E_{i,0}(\alpha) = \frac{\Phi_{i}^- \chi \lambda_{i,0}(\alpha)}{[\lambda_{i,0}^2 + \Phi_{i}^+ \Phi_{i}^- \chi^2 + \Phi_{i}^+ \Phi_{i}^- \chi^2]}\).

At \(\chi_{bs} = 0\), \(E_{i,0}(\alpha)\) becomes \(E_{i}\) and \(C_{i,0}(\alpha)\) becomes \(C_i(\alpha) = 1 - E_i(\alpha)\).

B. Steady currents

The steady current is given by \(I_{bs}^{\text{Steady}}(\alpha) = \frac{\partial \lambda_{i,0}(\alpha)}{\partial (\chi_{bs})} \big|_{\chi = 0}\). In non-interacting case, \(\lambda_{i,0}(\alpha) = \sum_i \lambda_{i,0}(\alpha)\) and it leads to \(I_{bs}^{\text{Steady}}(\alpha) = \sum_i I_{bs}^{\text{Steady}}(\alpha)\). Here, \(I_{bs}^{\text{Steady}}(\alpha) = \frac{\partial \lambda_{i,0}(\alpha)}{\partial (\chi_{bs})} \big|_{\chi = 0}\) are calculated from Eq.\((41)\) as

\[
I_{bs}^{\text{Steady}}(\alpha) = \frac{\Gamma_{b,s} \Gamma_{R,s} (f_R(\omega_i) - f_L(\omega_i))}{\Gamma_i}, \tag{48}
\]

with \(\Gamma_{b,s} = 2\pi \sum_{k} |W_{bks,i}|^2 \delta(\epsilon_{bk} + \sigma g_b B_b - \omega_i), \Gamma_{b,i} = \sum_{k} \Gamma_{b,s} \Gamma_{b,s} \Gamma_{i,s} \Gamma_{i,s} \). \(\Gamma_{bs}^{\text{Steady}}(\alpha)\) vanishes at zero bias \((\beta_b = 0, \mu_b = \mu_R)\).

Let us consider the modulation of only the thermodynamic parameters \(\alpha' = \{\mu_b, \chi_b\}_{b=L,R}\) similarly to Refs.\[32,37,38,42\]. The factor depending on \(\alpha'_{bs}^{\text{Steady}}(\alpha)\) is \((f_{\beta_b(t),\mu_b(t)}(\omega) - f_{\beta_L(t),\mu_L(t)}(\omega))\) with \(f_{\beta,\mu}(\omega) = [e^{i\beta(\omega - \mu)} + 1]^{-1}\). Hence,

\[
\langle \Delta N_{bs} \rangle_{\tau}^{\text{Steady}} = \sum_i \frac{\Gamma_{b,s} \Gamma_{R,s} \Gamma_{R,s} \Gamma_{i,s}}{\Gamma_i} \times \int dt [f_{\beta_b(t),\mu_b(t)}(\omega) - f_{\beta_L(t),\mu_L(t)}(\omega)] \tag{49}
\]

is generally nonzero and much longer than \(\langle \Delta N_{bs} \rangle_{\tau}^{\text{Berry}}\) because the period \(\tau\) is large for adiabatic pumping. Similarly, we can show that \(\langle \Delta N_{bs} \rangle_{\tau}^{\text{Steady}}\) is generally nonzero for interacting system \(\{V\oplus B\}\).

References\[28,29\] chose \(V = \mu_L - \mu_R\) as one of the modulating parameters, which also makes \(\langle \Delta N_{bs} \rangle_{\tau}^{\text{Steady}}\) to be nonzero. References\[32,37,38,42\] considered special modulations of only thermodynamic parameters which satisfy \(\langle \Delta N_{bs} \rangle_{\tau}^{\text{Steady}} = 0\). In fact, the steady currents are always zero for arbitrary modulations of only the dynamics parameters at zero-bias. Hence, we should fix the thermodynamic parameters at zero-bias when we want to detect \(\langle \Delta N_{bs} \rangle_{\tau}^{\text{Berry}}\) easily.

C. BSN curvatures

In the following, we consider one level system of which Hamiltonian is Eq.\((35)\) at \(U = 0\). The steady state is given by \(|\rho_{s,0}(\alpha)\rangle = \otimes_{s=\uparrow,\downarrow} |\rho_{s,0}(\alpha)\rangle\) because the Liouvillian is described by a summation \((\hat{K}^X = \sum_{s=\uparrow,\downarrow} \hat{K})\). Similarly, the corresponding left eigenvalue is given by \(\langle \rho_{s,0}(\alpha) | = \otimes_{s=\uparrow,\downarrow} \langle \rho_{s,0}(\alpha) |\). The BSN vectors Eq.\((11)\) are given by

\[
A_{bs,\alpha'}(\alpha) = \sum_{s=\uparrow,\downarrow} \nu_{s,0}(\alpha') \frac{\partial E_{s}(\alpha)}{\partial \alpha'}, \tag{50}
\]
where
\[ \psi_{s}^{b\sigma}(\alpha') = \frac{\partial \psi_{s}(\alpha)}{\partial (i\chi_{b\sigma})} \bigg|_{\chi=0} = \Gamma_{b\sigma,s} \alpha, \] (51)
with
\[ \Gamma_{b\sigma,s} = 2\pi \Delta_{b} \sum_{k} |v_{b\sigma,s}|^2 \times \delta (\varepsilon_{bk} + \sigma g_{b} B - \omega_{0} - sg_{S} B_{S}). \]
\(\psi_{s}^{b\sigma}(\alpha')\) does not depend on \(\alpha''\). Eq. [50] leads to an expression of the BSN curvatures
\[ F_{mn}^{b\sigma}(\alpha) = \sum_{s=\uparrow,\downarrow} \left[ \frac{\partial \psi_{s}^{b\sigma}(\alpha')}{\partial \alpha^m} \cdot \partial E_{s}(\alpha) - (m \leftrightarrow n) \right]. \] (52)
If \(\alpha^{m}\) or \(\alpha^{n}\) are elements of \(\alpha''\), \(F_{mn}^{b\sigma}(\alpha) = 0\). This is consistent with the result of Ref. [32]. In the following, we fix \(\alpha''\) to zero-bias (\(\beta_{b} = \beta, \mu_{b} = \mu\)). In this case, \(E_{s}(\alpha)\) is given by \(E_{s}(\alpha) = f(\omega_{0} + sg_{S} B_{S})\) with \(f(\omega) = [e^{\beta(\omega - \mu)} + 1]^{-1}\).

Now we approximate the linewidth as
\[ \Gamma_{b\sigma,s} = \delta_{\sigma,s}[\Gamma_{b} + \Gamma_{b}' (sg_{S} B_{S} - \sigma g_{b} B_{b})] \]
\[ = \delta_{\sigma,s} \Delta_{b} [\gamma_{b} + \gamma_{b}' (sg_{S} B_{S} - \sigma g_{b} B_{b})], \]
where \(\Gamma_{b}'\) are energy differential coefficients of linewidth functions at \(B_{b} = B_{S} = 0\). Namely, we disregard spin flips induced by tunneling between the dot and the leads are negligible and the magnetic fields \(B_{S}, B_{b}\) are small enough that \(|\Gamma_{b}'(sg_{S} B_{S} - g_{b} B_{b})| \ll \Gamma_{b}\) are satisfied. In this condition, \((\alpha^{m}, \alpha^{n}) = (B_{L}, B_{S}), (\Delta_{L}, B_{S})\) components of the charge and spin BSN curvatures of lead \(L\) are

\[
F_{BL,BS}^{L_{1}} \pm F_{BL,BS}^{L_{1}} = g s g_{L} \Gamma_{L}[f'(\omega_{0} + g s B_{S}) \pm f'(\omega_{0} - g s B_{S})] \left( \frac{\Gamma_{R}}{\Gamma_{L}^{2}} \right) \\
+ g s g_{L} \Gamma_{L}[f'(\omega_{0} + g s B_{S}) \mp f'(\omega_{0} - g s B_{S})] \left( \frac{\Gamma_{R}}{\Gamma_{L}^{0}} \right) \\
\times \left( \Gamma_{L} \frac{2 \Gamma_{R}^{2}}{\Gamma_{S}^{2}} + \Gamma_{R}\right) \left( g s B_{S} - g_{L} B_{L} \right) \frac{\Gamma_{R} - \Gamma_{L}}{\Gamma_{L}^{3}}. \] (53)

\[
F_{BL,BS}^{L_{1}} \pm F_{BL,BS}^{L_{1}} = g s [f'(\omega_{0} + g s B_{S}) \mp f'(\omega_{0} - g s B_{S})] \left( \frac{\gamma_{L} R \Delta_{R}}{\gamma_{L} \Delta_{L} + \gamma_{R} \Delta_{R}} \right)^{2} \\
\times g s [f'(\omega_{0} + g s B_{S}) \pm f'(\omega_{0} - g s B_{S})] \left( g s B_{S} - g_{L} B_{L} \right) \frac{\gamma_{R} R \Delta_{R} - \gamma_{L} \Delta_{L}}{\gamma_{L} \Delta_{L} + \gamma_{R} \Delta_{R}}^{2}. \] (54)

V. INTERACTING SYSTEM

In this section, we study the interacting system Eq. (35). First, we explain the Liouvillian for \(0 \leq U \leq \infty\) ((V A). Next, the steady charge and spin currents are calculated at \(U = \infty\) ((V B). Finally, the BSN curvatures corresponding to Eq. (35) and Eq. (54) are calculated at \(U = \infty\) ((V C) and differences of the results between \(U = 0\) and \(U = \infty\) are discussed.

A. Liouvillian

We explain the Liouvillian for \(k_{B} T > \Gamma\). In the Kondo region, the Born-Markov approximation is not good. However, for higher temperature region \((k_{B} T > \Gamma)\) this approximation is appropriate, where the Kondo effect is not important.

The matrix representation of the Liouvillian of RWA by the number states \(\{|n_{L},n_{R}\}\} (n_{s} = 0, 1\) are the number of an electron with spin \(s = \uparrow, \downarrow\) is a \(16 \times 16\)-matrix which is block diagonalized to the “diagonal” space (spanned
by \{ |n_1 n_2\rangle \langle n_1 n_2| \}_{n_1, n_2 = 0,1}\) and the “off-diagonal” space (spanned by \{ |n_1 n_4\rangle \langle m_1 m_4| \}_{(n_1, n_4) \neq (m_1, m_4)}\). The “diagonal” block is given by

\[
K^{\chi}(\alpha) = \begin{pmatrix}
-\Phi_\uparrow^+ + \Phi_\uparrow^- & \Phi_\uparrow^- - \Phi_\uparrow^+ \\
\Phi_\uparrow^+ - \Phi_\uparrow^- & -\Phi_\uparrow^+ + \Phi_\uparrow^-
\end{pmatrix}
\begin{pmatrix}
\Phi_\downarrow^+ - \Phi_\downarrow^- \\
-\Phi_\downarrow^- + \Phi_\downarrow^+
\end{pmatrix}
\begin{pmatrix}
0 \\
0
\end{pmatrix}
\begin{pmatrix}
|0000\rangle \\
|1010\rangle
\end{pmatrix},
\]

with

\[
\phi_{b,s}^{\pm} = 2\pi \Delta_b \sum_{k,\sigma} \langle v_{b\sigma,s} | f_{b^\pm}(\omega_0 + s g s B_s + U) \times e^{\mp i \Delta_b} \delta_{kk} + s g \phi B_s - \omega_0 - s g s B_s - U, \rangle
\]

and \( \phi_{b,s}^{\pm} = \phi_{b,s}^{\pm} |_{U=0} \). The “off-diagonal” block is a \(12 \times 12\)-diagonal matrix, which does not relate to the steady state. At \( U = 0, K^{\chi}(\alpha) \) becomes \( K^\chi(\alpha) \otimes 1_{1\uparrow} + 1_{\downarrow} \otimes K^\chi(\alpha) \), where \( K^\chi(\alpha) \) is identity matrices. In the opposite limit \( U \to \infty, K^\chi(\alpha) \) reduces to

\[
K^{\chi(\infty)}(\alpha) = \begin{pmatrix}
-\Phi_\uparrow^+ + \Phi_\uparrow^- & \Phi_\uparrow^- - \Phi_\uparrow^+ \\
\Phi_\uparrow^+ - \Phi_\uparrow^- & -\Phi_\uparrow^+ + \Phi_\uparrow^-
\end{pmatrix}
\begin{pmatrix}
\Phi_\downarrow^+ - \Phi_\downarrow^- \\
-\Phi_\downarrow^- + \Phi_\downarrow^+
\end{pmatrix}
\begin{pmatrix}
0 \\
0
\end{pmatrix}
\begin{pmatrix}
|0000\rangle \\
|1010\rangle
\end{pmatrix},
\]

because the density of state of both leads vanishes at high energy (\( \phi_{c,s}^{\pm} \to 0 \)).

### B. Steady currents

In this section, we set \( U = \infty \). The characteristic polynomial of \( K^{\chi(\infty)} \) is denoted as \( C_3(\chi; \lambda) = \text{det}(K^{\chi(\infty)} - \lambda) = \sum_{n=0}^2 C_n(\chi) \chi^n - \lambda^3 \). Because of \( C_0(0) = 0, \lambda = 0 \) is one of the solutions at \( \chi = 0 \). Now we set \( \chi_{bs} \) is infinitesimal and other counting fields are zero. Then, the eigenvalue corresponding to the steady state is given by \( \lambda = \lambda_0(\alpha) = i \chi_{bs} \cdot \rangle \langle \text{Steady} \rangle + \mathcal{O}(\chi_{bs}^2) \). It leads to

\[
0 = C_3(\chi; \lambda_0(\alpha)) = C_1(0) i \chi_{bs} \cdot \rangle \langle \text{Steady} \rangle + i \chi_{bs} C_0 \]

with \( C_0 = \frac{\partial C_3(\chi)}{\partial \chi_{bs}} |_{\chi=0} \), and we obtain

\[
I^{\text{Steady}}_{b\sigma} = -C_{b\sigma}^{\text{Steady}} |_{\chi=0},
\]

with \( C_1(0) = -[\Phi_\uparrow^+ \Phi_\uparrow^- + \Phi_\uparrow^- \Phi_\uparrow^+] |_{\chi=0} \). From \( C_0(0) = -[\Phi_\uparrow^+ \Phi_\uparrow^- + \Phi_\uparrow^- \Phi_\uparrow^+] |_{\chi=0} \), we have

\[
I^{\text{Steady}}_{b\sigma}(\alpha) = \sum_{s=\uparrow, \downarrow} \frac{\Phi_\uparrow^+ + \Phi_\uparrow^-}{\Phi_\uparrow^+ - \Phi_\uparrow^-} \Gamma_{L,s} \Gamma_{R,s} (f_{R,s} - f_{L,s})
\]

where \( \uparrow = \downarrow, \downarrow = \uparrow \). At zero-bias, the steady currents vanish. Similar to (\ref{eq:dNSteady}) \( \langle \Delta N_{b\sigma} \rangle^{\text{Steady}} \rangle \) is generally nonzero when \( \alpha'' \) is not fixed at zero-bias.

### C. BSN curvatures

The steady state \( \rho_0(\alpha) \) and corresponding left eigenvector \( l_0^\sigma(\alpha) \) are written as

\[
\rho_0(\alpha) = \rho_0 |00\rangle \langle 00| + \rho_1 |10\rangle \langle 10| + \rho_2 |01\rangle \langle 01| + \rho_1 |11\rangle \langle 11| + \rho_2 |00\rangle \langle 00| + l_0^\sigma |10\rangle \langle 10| + l_0^\sigma |01\rangle \langle 01| + l_0^\sigma |11\rangle \langle 11|.
\]

The BSN vectors are given by

\[
A_n^{b\sigma}(\alpha) = \sum_{c=\uparrow, \downarrow} l_{b\sigma}^{c}(\alpha) \frac{\partial \rho_c(\alpha)}{\partial \alpha_n},
\]

where \( l_{b\sigma}^{c}(\alpha) = \frac{\partial \rho_c(\alpha)}{\partial \alpha_n} |_{\chi=0} \). It leads to the BSN curvatures

\[
F_{mn}^{b\sigma}(\alpha) = \sum_{c=\uparrow, \downarrow} \frac{\partial l_{b\sigma}^{c}(\alpha)}{\partial \alpha_m} \frac{\partial \rho_c(\alpha)}{\partial \alpha_n} - (m \leftrightarrow n)
\]

Particularly, in the \( U \to \infty \) limit, \( \rho_2 \) vanishes and \( F_{mn}^{b\sigma}(\alpha) \) reduces to

\[
F_{mn}^{b\sigma(\infty)}(\alpha) = \sum_{s=\uparrow, \downarrow} \frac{\partial l_{b\sigma}^{s}(\alpha)}{\partial \alpha_m} \frac{\partial \rho_s^{\infty}(\alpha)}{\partial \alpha_n} - (m \leftrightarrow n),
\]

where \( \rho_s^{\infty}(\alpha) \) and \( l_{b\sigma}^{s}(\alpha) \) are the limits in \( U \to \infty \) of \( \rho_s(\alpha) \) and \( l_{b\sigma}^{s}(\alpha) \), respectively. From Eq.\ref{eq:s} we obtain

\[
\rho_s^{\infty}(\alpha) = \frac{\Phi_\uparrow^+ \Phi_\downarrow^-}{\Phi_\uparrow^+ \Phi_\downarrow^- + \Phi_\uparrow^+ \Phi_\uparrow^- + \Phi_\downarrow^+ \Phi_\downarrow^-}.
\]

\[
l_{b\sigma}^{s}(\alpha) = \frac{\Phi_\uparrow^+ \Phi_\downarrow^-}{\Phi_\uparrow^+ \Phi_\downarrow^- + \Phi_\uparrow^+ \Phi_\uparrow^- + \Phi_\downarrow^+ \Phi_\downarrow^-},
\]

with \( \omega_s = \omega_0 + g s B_s \). In the following, we fix \( \alpha'' \) to zero-bias (\( \beta_0 = \beta, \mu_0 = \mu \)). Then, \( l_{b\sigma}^{s}(\alpha^{\infty}) \) equals to \( V_{bs}(\alpha^{s}) \) given by Eq.\ref{eq:51} and \( \rho_s^{\infty}(\alpha) \) are given by \( \rho(s B_S) \) with

\[
\rho(s B_S) = \frac{e^{-\beta(\omega_0 - \mu)}}{1 + e^{-\beta(\omega_0 - \mu)}} + e^{-\beta(\omega_0 - \mu)}.
\]

Eq.\ref{eq:61} can be obtained by a replacement

\[
E_s(\alpha) = f(\omega) \to \rho(s B_S)
\]
in Eq. (2). The charge and spin BSN curvatures of \((B_L, B_S), (\Delta_L, B_S)\)-pump are given by a replacement \(f'(\pm g_S B_S) \rightarrow \rho'(\pm B_S)\) in Eqs. (3) and (4), where \(\rho'(B_S) = \frac{1}{g_S} \partial \rho(B_S) / \partial B_S\). Similarly to \(U = 0\), the charge and spin BSN curvatures of \((B_L, B_R), (\Delta_L, \Delta_R)\)-pump are zero.

In Figs. (a)-(d), we plot the BSN curvatures of \((B_L, B_S)\)-pump times \((-1)\). For \(U = 0\), the charge and spin BSN curvatures are shown in Fig. (a) and Fig. (b), and for \(U = \infty\) these are shown in Figs. (c),(d). The vertical and horizontal axes of these plots are the strength of magnetic fields \(B_S, B_L\) normalized by \(\Gamma / \mu_B\), where \(\Gamma = \Gamma_L = \Gamma_R\) and \(\mu_B = 57.88 \, \mu eV / \Gamma\) is the Bohr magneton. The values of the parameters used for these plots are \(\gamma_L = \gamma_R = \Gamma, \gamma'_L = \gamma'_R = 0.1, \beta = 0.5 / \Gamma, \omega_0 = \mu - 3 \Gamma, B_R = 0\), and \(g^L_S = g^R_S = -0.44\) (bulk GaAs). The BSN curvatures of \((\Delta_L, B_S)\)-pump are shown similarly in Figs. (a)-(d). In all plots, \(\gamma_L = \Gamma_R = \Gamma, \gamma'_L = \gamma'_R = 0.1\), and other conditions are the same as in Fig. (a). The first and second terms of the right side of Eq. (3) are the second and third order of \(g\)-factor, and the first and second terms of the right side of Eq. (4) are the first and second order of \(g\)-factor. If all \(g\)-factors change to \(-20\) (for example for the materials like InAs, InSb), the 1st, 2nd, 3rd order terms become about 45, 2 000, 90 000 times.

Figure (a) shows the average number of up spin electrons of the quantum dot \(F(B_S) = f(\omega_0 + g_S B_S)\) for \(U = 0\) (solid line), \(\rho(B_S)\) for \(U = \infty\), dashed line) for \(\beta = 0.5 / \Gamma, \omega_0 = \mu - 3 \Gamma\) and \(g_S = -0.44 \times \mu_B / 2\). Because two electrons cannot occupy a quantum dot at \(U = \infty\), the magnetic filed dependence of \(\rho(B_S)\) is more sensitive than \(f(\omega_0 + g_S B_S)\). Figures (b), (c) show \(F'(\pm B_S) \mp F'(-B_S)\) normalized by \(\Gamma / \mu_B\), where \(F'(\pm B_S) = \frac{1}{g_S} \partial F(B_S) / \partial B_S\) at \(B = \pm B_S\).

In \(\omega_0 > \mu\) region, the larger \(\omega_0 - \mu\), the less the difference between \(U = 0\) and \(U = \infty\) becomes. The Coulomb interaction prevents that two electrons occupy the quantum dot. This effect is conspicuous in \(\omega_0 < \mu\) region although it is not important in \(\omega_0 > \mu\) region.

As shown in Figs. (a),(c) and Figs. (b),(d), the \(B_S\)-dependence of the charge BSN curvature of \((B_L, B_S)\)-pump and the spin BSN curvature of \((\Delta_L, B_S)\)-pump at
FIG. 2: (Color online) (a) The BSN curvature of charge of $(\Delta L, B_S)$-pump, $-[F^y_{\Delta L, B_S} + F_{\Delta L, B_S}]$ at $U = 0$, (b) the BSN curvature of spin, $-[F^y_{\Delta L, B_S} - F_{\Delta L, B_S}]$ at $U = 0$, (c) $-[F^y_{\Delta L, B_S} + F_{\Delta L, B_S}]$ at $U = \infty$, (d) $-[F^y_{\Delta L, B_S} - F_{\Delta L, B_S}]$ at $U = \infty$. The values of the parameters used for these plots are $\gamma_L = \Gamma_R = \Gamma$, $\gamma'_L = \Gamma'_R = 0$, and other conditions are same as Fig. 1.

FIG. 3: (Color online) (a) $F(B_S) = f(\omega_0 + g_S B_S)$ (solid line), $\rho(B_S)$ (dashed line). (b) $F'(B_S) - F'(-B_S)$, (b) $F'(B_S) + F'(-B_S)$, where $F'(B_S) = \frac{1}{g_S} \frac{\partial F(B_S)}{\partial B}|_{B=\pm B_S}$. In all plots, $\beta = 0.5/\Gamma$, $\omega_0 = \mu - 3\Gamma$ and $g_S = -0.44 \times \mu_B/2$ with the Bohr magneton $\mu_B = 57.88 \mu eV/T$.

$U = 0$ are more gentle than those at $U = \infty$. It results from the behavior of $F'(B_S) + F'(-B_S)$ as shown in Fig. 3(c).

As shown in Fig. 1(b),(d) and in Figs. 2(a),(c), the $B_S$-dependence of the spin BSN curvature of $(B_L, B_S)$-pump and the charge BSN curvature of $(\Delta L, B_S)$-pump are opposite. This is because the leading term (in weak magnetic field region) of these are proportional to $F'(B_S) - F'(-B_S)$ and its $B_S$-dependence is opposite in $U = 0$ and $U = \infty$ for $\omega_0 - \mu < 0$ as indicated in Fig. 3(b). This inversion is realized for only $\omega_0 - \mu < 0$ region. At $\omega_0 = \mu$, $f'(\omega_0 + g_S B_S) - f'(\omega_0 - g_S B_S)$ vanish. In $\omega_0 > \mu$
region, the signs of $f'(\omega_0 + gSB_S) - f'(\omega_0 - gSB_S)$ and $\rho'(B_S) - \rho'(-B_S)$ are the same.

VI. CONCLUSION

In this paper, we investigated quantum adiabatic pump of charge and spin using the GQME (generalized quantum master equation) approach proposed by Yuge et al.\textsuperscript{22} In \textsuperscript{111} we studied non-adiabatic effect and showed the correspondence between our approach and the real-time diagrammatic approach\textsuperscript{20}. Moreover, we showed that the Berry-Sinitsyn-Nemenman (BSN) phase derived under the adiabatic condition has the non-adiabatic information. The GQME picks out one higher order non-adiabatic information from the solution of the QME. Particularly, the steady state (the zeroth order of the pumping frequency) gives first order response (pumped current). This fact may be related to Ref.\textsuperscript{13} which is connected the BSN vector and the linear response theory of the QME.

We generalized the GQME approach to multi-counting field (\textsuperscript{11A}) and studied quantum dots system weakly coupled to leads ($L$ and $R$) in \textsuperscript{11V} and \textsuperscript{11VI} We showed that the pumped charge and spin coming from the steady current are not negligible when the thermodynamic parameters (the chemical potentials and the temperatures of leads) are not fixed to zero-bias in non-interacting quantum dots (\textsuperscript{11VII}) and an interacting dot (\textsuperscript{11VIII}). We focused on the dynamic parameters (the magnetic fields applied collinear to quantum dots ($B_S$) and leads ($B_L$, $B_R$) and the coupling strength between dots and leads, $\Delta_{L/R}$) as control parameters.

In one level system with the Coulomb interacting $U$, we analytically calculated the BSN curvatures of spin and charge of ($B_L$, $B_S$)-pump and ($\Delta_L$, $B_S$)-pump for the non-interacting limit ($U = 0$) and the strong interaction limit ($U = \infty$) using the rotating wave approximation (RWA) defined as the long coarse-graining time limit of the coarse-graining approximation (CGA). The difference between $U = 0$ and $U = \infty$ appeared through the factor denoting the average number of up or down spin electrons of the quantum dot. Additionally, the adiabatic modulations of ($B_L$, $B_R$) or ($\Delta_L$, $\Delta_R$) cannot pump both charge and spin.

In this paper, only $U = 0$ and $U = \infty$ limits are studied. But we can analyze finite $U$ based on Eq.\textsuperscript{60}. This is our future work. Recently, R. Yoshii and H. Hayakawa\textsuperscript{22} studied adiabatic pump of charge by only the thermodynamic parameters using the same approach in a similar system without magnetic fields (for finite $U$). The work (for the thermodynamic parameters) involving the above problem is contrastive to our work (for the dynamic parameters).

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Appendix A: Liouville space

By following correspondence, an arbitrary linear operator (which operates to the Hilbert space) $\bullet = \sum_{n,m} \langle n| \bullet |m\rangle \langle m| \langle n|$ is mapped to a vector of the Liouville space\textsuperscript{21,39} $\bullet) = \sum_{n,m} \langle n| \bullet |m\rangle |nm\rangle$:

$$|n\rangle\langle m| \quad \leftrightarrow \quad |nm\rangle, \quad (A1)$$
$$\text{Tr}(|m\rangle\langle n'|\langle m'|) \quad \leftrightarrow \quad \langle nm|n'm'\rangle, \quad (A2)$$
$$\text{Tr}(A^{\dagger}B) \quad \leftrightarrow \quad \langle A|B\rangle, \quad (A3)$$
$$\text{Tr}(\bullet) \quad \leftrightarrow \quad \langle 1|\bullet\rangle. \quad (A4)$$

Here, $\{\{n\}\}$ is an arbitrary complete orthonormal basis. The inner product of the Liouville space is defined by the Hilbert-Schmidt product (Eq.(A3)). The Hermitian conjugate of $\bullet$ is defined as $\langle\bullet\rangle = (\{\bullet\})^{\dagger} = \sum_{n,m} \langle n| \bullet |m\rangle^{\dagger} \langle nm\rangle$. An arbitrary linear super-operator $\hat{J}$ which operates to any operator $\bullet$ is mapped to a corresponding operator of the Liouville space ($\hat{J}$) as

$$|\hat{J}\bullet\rangle = \hat{J}|\bullet\rangle. \quad (A5)$$

The matrix representation of $\hat{J}$ (or $\hat{J}$) is defined by

$$J_{nm,kl} = \langle nm|\hat{J}|kl\rangle. \quad (A6)$$

In the main text of this paper, both $\hat{J}$ and $\hat{J}$ are denoted by $\hat{J}$.

Generally, the Liouvillian $\hat{K}$ operates to an operator $\bullet$ as

$$\hat{K}\bullet = -i[H_S, \bullet] + \hat{P}\bullet, \quad (A7)$$
$$\hat{P}\bullet = \sum_{a} A_{a}\bullet B_{a}, \quad (A8)$$

where $H_S$ is a Hermitian operator and $A_{a}$, $B_{a}$ are operators. The matrix representation of Eq.(A7) is given by

$$\sum_{k,l} K_{nm,kl} \bullet_{kl} = \sum_{k,l} \left[ -i \{(H_S)_{nk}\delta_{lm} - \delta_{nk}(H_S)_{lm}\} \bullet_{kl} \right. \right.$$}
$$+ \left. \left[ \sum_{a} (A_{a})_{nk}(B_{a})_{lm}\right] \bullet_{kl} \right]. \quad (A9)$$

where $\bullet_{kl} = \langle k|\bullet|l\rangle$. Hence, the matrix representation of $\hat{K}$ is given by

$$K_{nm,kl} = -iH_{nm,kl} + \Pi_{nm,kl}, \quad (A10)$$
$$H_{nm,kl} = (H_S)_{nk}\delta_{lm} - \delta_{nk}(H_S)_{lm}, \quad (A11)$$
$$\Pi_{nm,kl} = \sum_{a} (A_{a})_{nk}(B_{a})_{lm}. \quad (A12)$$
Appendix B: Coarse-graining approximation

\[ \rho_{\text{tot}}(t)(\Omega) \] is governed by modulated von Neumann equation\(^{3} \):

\[ \frac{d}{dt}\rho_{\text{tot}}(t) = -i[H_{\text{tot}}(t), \rho_{\text{tot}}(t)]_{\chi}, \quad (B1) \]

where \( H_{\text{tot}}(t) \) is the total Hamiltonian and \([A, B]_{\chi} = A_{\chi}B - BA_{\chi} \) with \( A_{\chi} = e^{i\sum_x \chi_x \hat{O}_x/2}A e^{-i\sum_x \chi_x \hat{O}_x/2} \).

\( H_{\text{tot}}(t) \) is given by \( H_{\text{tot}}(t) = H_{S}(t) + \sum_b[H_b(t) + H_{1b}(t)] \),

where \( H_{S} \) is the system (denoted by \( S \)) Hamiltonian, \( H_b \) is the Hamiltonian of bath \( b \), and \( H_{1b} \) is the tunneling interaction Hamiltonian between \( S \) and bath \( b \). In the following, we suppose Eq.(35), Eq.(36), Eq.(37) for arbitrary number of leads \( (b = 1, 2, \cdots, M) \).

Now we move to the interacting picture. An operator in the interacting picture corresponding to \( A(t) \) is defined by \( A^{I}(t) = U_{0}^\dagger(t)A(t)U_{0}(t) \) with \( \frac{dU_{0}(t)}{dt} = -i[H_{S}(t) + \sum_b[H_b(t) + H_{1b}(t)] \) and \( U_{0}(0) = 1 \). The system reduced density operator in the interacting picture is given by \( \rho^{I,\chi}(t) = \text{Tr}_{\text{leads}}[\rho_{\text{tot}}^{I}(t)] \) where \( \rho_{\text{tot}}^{I}(t) = U_{0}(t)\rho(t)U_{0}^\dagger(t) \) and \( \text{Tr}_{\text{leads}} \) denotes the trace of leads space. Up to the second order perturbation about \( H_{1} = \sum_b H_{1b} \), we obtain

\[ \rho^{I,\chi}(t + T) = \rho^{I,\chi}(t) - \int_{t}^{t+T} du \int_{u}^{T} ds \text{Tr}_{\text{leads}}\left[[H_{1}^{I}(u), [H_{1}^{I}(s), \rho^{I,\chi}(t)\rho_{B}(\alpha_{t})]]_{\chi}\right] \]

\[ \equiv \rho^{I,\chi}(t) + T\hat{L}_{\chi}^{I}(t)\rho^{I,\chi}(t), \quad (B2) \]

for an arbitrary \( T \geq 0 \). Here, we use the Born-Markov approximation \( \rho_{\text{tot}}^{I,\chi}(t) \approx \rho^{I,\chi}(t)\rho_{B}(\alpha_{t}) \), where \( \rho_{B}(\alpha_{t}) = \bigotimes_{b} e^{-\beta_{b}(t)[H_b(t) - \mu_{b}(t)N_{b}]/\Xi_{b}(\alpha_{t})} \) with \( \Xi_{b}(\alpha_{t}) = \text{Tr}_{b}[e^{-\beta_{b}(t)[H_b(t) - \mu_{b}(t)N_{b}]}] \) and \( \text{Tr}_{b} \) denotes the trace of lead \( b \). The coarse-graining approximation\(^{3,35,36} \) (CGA) is defined by

\[ \frac{d}{dt}\rho^{I,\chi}(t) = \hat{L}_{\chi}^{I}(t)\rho^{I,\chi}(t). \quad (B3) \]

If the cycle time of the modulation of control parameters \( (\tau) \) is much larger than the coarse-graining time \( T \), the super-operator \( \hat{L}_{\chi}^{I}(t) \) is described as a function of the set of control parameters at time \( t \) \( (\alpha_{t}) \). In this paper, we suppose \( \tau \gg T \).

In the Schrödinger picture, Eq.(B3) is described as

\[ \frac{d\rho^{I,\chi}(t)}{dt} = -i[H_{S}(t), \rho^{I,\chi}(t)] + \hat{\Pi}^{I}_{\chi}(\alpha_{t})\rho^{I,\chi}(t). \quad (B4) \]

Here, the super-operator \( \hat{\Pi}^{I}_{\chi}(\alpha) \) operates to an operator \( \bullet \) as

\[ \hat{\Pi}^{I}_{\chi} \bullet \rightarrow \sum_{b} \sum_{\omega,\omega',n,m,s,s'} \left[ \Phi_{b,n,s,m,s'}^{\pm}(\chi, T, \omega, \omega')a_{m,s'}(\omega') \bullet a_{n,s}(\omega) - a_{n,s}(\omega) \bullet a_{m,s'}(\omega') \right] \]

\[ -\frac{1}{2}\Phi_{b,n,s,m,s'}^{\pm}(T, \omega, \omega')a_{n,s}(\omega) a_{m,s'}(\omega') \bullet a_{n,s}(\omega) + \Phi_{b,n,s,m,s'}^{\mp}(\chi, T, \omega, \omega') a_{m,s'}(\omega') \bullet a_{n,s}(\omega) \]

\[ -\frac{1}{2}\Phi_{b,n,s,m,s'}^{\mp}(T, \omega, \omega') a_{n,s}(\omega) a_{m,s'}(\omega') \bullet a_{n,s}(\omega) + \frac{1}{2}\Phi_{b,n,s,m,s'}^{\pm}(T, \omega, \omega') a_{m,s'}(\omega') a_{n,s}(\omega) \bullet a_{n,s}(\omega) \]. \quad (B5) \]

where

\[ X^{\pm}(\chi, T, \omega, \omega') = \frac{e^{\pm i(\omega - \omega') \chi}}{2\pi} \int_{-\infty}^{\infty} d\Omega X^{\pm}(\Omega, \chi) T \text{sinc} \left( \frac{T(\Omega - \omega)}{2} \right) \text{sinc} \left( \frac{T(\Omega - \omega')}{2} \right), \quad (B6) \]

and \( X^{\pm}(T, \omega, \omega') = X^{\pm}(\chi = 0, T, \omega, \omega') \). Here, \( \text{sinc}(x) = \sin x/x \) and \( X^{\pm}(\Omega, \chi) \) denotes one of \( \Phi_{b,n,s,m,s'}^{\pm}(\Omega, \chi), \)
The elements of $\Psi_{b,n,s,m}(\Omega, \chi)$, where

\begin{align}
\Phi_{b,n,s,m}(\Omega, \chi) &= 2\pi \sum_{k,\sigma} V_{b,k,\sigma} V_{b,k,\sigma}^* \left[ 1 - f_b(\varepsilon_{bk\sigma}) \right] e^{i\chi_{b\sigma}} e^{i\lambda_{b\sigma}} \delta(\varepsilon_{bk\sigma} - \Omega), \\
\Phi_{b,n,s,m}(\Omega, \chi) &= 2\pi \sum_{k,\sigma} V_{b,k,\sigma} V_{b,k,\sigma}^* \left[ 1 - f_b(\varepsilon_{bk\sigma}) \right] e^{-i\chi_{b\sigma}} e^{-i\lambda_{b\sigma}} \delta(\varepsilon_{bk\sigma} - \Omega), \\
\Psi_{b,n,s,m}(\Omega, \chi) &= 2\pi \sum_{k,\sigma} V_{b,k,\sigma} V_{b,k,\sigma}^* \left[ 1 - f_b(\varepsilon_{bk\sigma}) \right] e^{i\chi_{b\sigma}} e^{i\lambda_{b\sigma}} P \frac{1}{\varepsilon_{bk\sigma} - \Omega}, \\
\Psi_{b,n,s,m}(\Omega, \chi) &= 2\pi \sum_{k,\sigma} V_{b,k,\sigma} V_{b,k,\sigma}^* \left[ 1 - f_b(\varepsilon_{bk\sigma}) \right] e^{-i\chi_{b\sigma}} e^{-i\lambda_{b\sigma}} P \frac{1}{\varepsilon_{bk\sigma} - \Omega},
\end{align}

with $V_{b,k,\sigma} = \sqrt{\Delta b} c_{b\sigma}$, $\varepsilon_{b\sigma}$ and $\lambda_{b\sigma}$ denote the counting fields for $N_{b\sigma}$ and $\sum_k \varepsilon_{bk} c_{b\sigma}^\dagger c_{b\sigma}$. $f_b(\varepsilon) = \{ \exp(\beta_b(\mu_b - \varepsilon)) + 1 \}^{-1}$ is the Fermi distribution function and $P$ denotes the Cauchy principal value. The eigenvectors $a_{n\sigma}(\omega)$ are given by

$$a_{n\sigma}(\omega) = \sum_{\alpha,\beta} \delta_{\omega,\alpha,\beta} \omega | E_\beta \rangle \langle E_\alpha | a_{n\sigma} | E_\beta \rangle \langle E_\alpha |, \quad (B11)$$

with $\omega_{\alpha,\beta} = E_{\alpha} - E_{\beta}$ and $H_S | E_\alpha \rangle = E_\alpha | E_\alpha \rangle$. $\omega$ is one of the elements of $\{ \omega_{\alpha,\beta} | E_\alpha \langle a_{n\sigma} | E_\beta \rangle \neq 0 \ (n, \sigma) \}$.

In this paper, the rotating wave approximation (RWA) is defined as the limit of $T \to \infty$ of CGA. In this limit ($T, \max_{\omega, \omega'}(\omega - \omega') \gg 1$), $X^\pm(\chi, T, \omega, \omega') \approx X^\pm(\omega, \chi) \delta(\omega - \omega')$ is realized because of the fact that

$$\lim_{T \to \infty} T \frac{\sin T(\Omega - \omega)}{T^2} \frac{\sin T(\Omega - \omega')}{2} = 2\pi \delta(\omega - \omega').$$

If $H_S$ is time-independent, this RWA is equivalent to usual RWA.\textsuperscript{33}

The GQME of CGA (for finite $T$) is more difficult to analyze than that of RWA. For one level system Eq. (B8), the matrix representation of the Liouvillian of the RWA by the number states $\{|n_{\uparrow}n_{\downarrow}\rangle\}$ is block diagonalized to the “diagonal” part (spanned by $\{|n_{\uparrow}n_{\downarrow}\rangle \mid n_{\uparrow}, n_{\downarrow} = 0, 1\}$) and the “off-diagonal” part (spanned by $\{|n_{\uparrow}n_{\downarrow}\rangle \mid n_{\uparrow}, n_{\downarrow} \neq 0, 1\}$). The “diagonal” block is given by Eq. (55) and the “off-diagonal” block is a $12 \times 12$-diagonal matrix. But one of CGA has off diagonal components which permit transitions between “diagonal” and “off-diagonal” if spins can flip by tunneling $H_1$. Even if spins cannot flip, “off-diagonal” block are not diagonal. Particularly for $U = 0$, the Liouvillean does not reduce to a summation of one particle Liouvillean $\{\hat{K}_{\chi}(\alpha) \otimes \hat{1}_{\uparrow} + \hat{1}_{\uparrow} \otimes \hat{K}_{\chi}(\alpha)\}$. The study of differences between the RWA and the CGA is a future work.

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