Macroscopic quantum violation of the fluctuation-dissipation theorem in equilibrium

Kentaro Kubo,[1,2,*] Kenichi Asano,[3,†] and Akira Shimizu[1,2,‡]

[1]*Department of Basic Science, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan*
[2]*Komaba Institute for Science, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan*
[3]*Center for Education in Liberal Arts and Sciences, Osaka University, Toyonaka, Osaka 560-0043, Japan*

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We examine the Hall conductivity of a macroscopic two-dimensional quantum system and show that the observed quantities can sometimes violate the fluctuation dissipation theorem (FDT), even in the linear response (LR) regime infinitesimally close to equilibrium. At low temperature and in strong magnetic field, which are experimentally accessible, the violation can be by an order of magnitude larger than the Hall conductivity itself. We further generalize the results and obtain a necessary condition for such large-scale violation to happen. This violation is a genuine quantum phenomenon that appears on a macroscopic scale when the time-reversal symmetry is broken (by, e.g., a magnetic field). Our results are not only bound to the development of the fundamental issues of nonequilibrium physics, but the idea is also meaningful for practical applications, since the FDT is widely used for the estimation of noises from the LRs. We give an alternative formula evaluating precisely the fluctuations from the LRs even in the quantum systems with large FDT violation.

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I. INTRODUCTION

According to the fluctuation dissipation theorem (FDT), fluctuation in a thermal equilibrium state should agree with the linear response (LR) function times temperature $T = 1/\beta$ (we take $k_B = 1$) [1–12]. While the FDT was originally proposed on the dissipative components of LRs such as the diagonal conductivity [1–3], it was later suggested [13,14] and proved [8] in classical systems that the FDT holds also for the dissipationless components of LRs, such as the Hall conductivity. Consequently, the FDT is accepted as a universal relation that holds for *all* LRs in classical systems. This contrasts with the responses to strong external fields that drive the systems far away from equilibrium, for which such a relation does not hold in general [15–30].

The most important aspect of the FDT is that it connects the results of completely different and independent experiments [1–12]: The LR function is obtained by measuring the response in a nonequilibrium state, whereas the fluctuation is obtained by measuring the time correlation in an equilibrium state. According to the FDT, one can tell the magnitude of the equilibrium fluctuation by measuring the response function in nonequilibrium, and vise versa. This nontrivial aspect of the FDT is utilized widely, e.g., to estimate noises (fluctuations) from the responses in electric circuits [31–33], optical devices [33–35], and gravitational-wave detectors [36].

However, the validity of the FDT is nontrivial in quantum systems [8–10]. In "deriving" the FDT microscopically [4–7], it was implicitly assumed that the disturbance by measurement was negligible. Such an ideal measurement is possible only for classical systems, because quantum systems follow the uncertainty relations [37–42]. The best solution seems to assume a quantum measurement as *quasiclassical*, emulating the ideal classical one as closely as possible [9,10]. Then, a question arises: Does the FDT hold in quantum systems if quasiclassical measurements are made?

This fundamental question has recently been solved in Refs. [9,10]. It was shown rigorously that the FDT, as a relation between the results of two different experiments, is partially violated in quantum systems. Unlike obvious violations far from equilibrium [15–30], this violation occurs between equilibrium fluctuations and the LRs to infinitesimal external fields.

However, one might conjecture that the violation would occur only at high frequency, $\hbar\omega \gtrsim T$, for the following reason. According to Nyquist [3], a macroscopic phenomenology holds using the observed LR functions as given parameters, although their values may be determined by quantum effects. Then the remaining quantum correction for the fluctuation and the LR should be only about the equipartition law [3]. Since it breaks down only at $\hbar\omega \gtrsim T$, so should do the FDT. In systems with the time reversal symmetry, this is indeed the case, e.g., for the violation caused by the detectors that cannot measure zero-point fluctuations [43–51]. Practically, such a violation at high ω (e.g., $\omega/2\pi \gtrsim 20$ GHz at $T = 1$ K) is negligible in most applications [31–36] (except at very low T) because their operation frequencies are much lower.

By contrast, if the time-reversal symmetry is broken, the violation found in Refs. [9,10] occurs even at low frequency, $\hbar\omega \ll T$, for certain responses and even in the quasiclassical measurement by means of the heterodyning [51] or quantum nondemolition [52–56] detectors. However, since Refs. [9,10] did not estimate the *magnitude* of the violation in actual systems, one might still conjecture that it would be very small in macroscopic systems because it is a genuine quantum phenomenon that vanishes at $\hbar = 0$.

*kubo@as.c.u-tokyo.ac.jp
†asano@celas.osaka-u.ac.jp
‡shmz@as.c.u-tokyo.ac.jp

In this paper, we explicitly calculate the magnitude of the violation for the Hall conductivity σ_{xy} of a macroscopic quantum system. We show that the violation can be larger by an order of magnitude than $|\sigma_{xy}|$ even at $\hbar\omega \ll T$, in contrast to the above conjecture. We also derive a condition for such a macroscopic violation of the FDT in general quantum systems.

These results imply that the foundation of nonequilibrium physics needs to be updated even in the LR regime infinitesimally close to equilibrium. Furthermore, our results also have an impact on practical applications because the standard technique of estimating equilibrium noises of currents is to estimate them from the LR functions using the FDT [31–36], but wrong predictions are obtained about the 'real antisymmetric parts' (see Sec. II), such as the Hall conductivity. We will give the correct method for estimating fluctuations from the LRs in such a case.

II. FDT VIOLATION IN HALL EFFECT

Consider a macroscopic two-dimensional electron system (2DES) in the x-y plane and apply a uniform magnetic field with a flux density, $\boldsymbol{B} = (0, 0, B)$. This system has been studied extensively in the context of the quantum Hall effect (QHE) [57–72]. Here, we study the FDT between the conductivity tensor, $\sigma_{\mu\nu}$, and the time correlation $\Xi_{\mu\nu}$ of $\hat{j}_\mu := \hat{J}_\mu/\sqrt{\Omega}$ and $\hat{j}_\nu := \hat{J}_\nu/\sqrt{\Omega}$, where $\hat{J}_\mu$ ($\mu = x, y$) is the total current and Ω is the area of the system. For simplicity, we assume that the system is invariant under rotation by $\pi/2$ about the z axis. Then, the Hall conductivity has only the antisymmetric part, $\sigma_{xy} = \sigma^-_{xy} := (\sigma_{xy} - \sigma_{yx})/2$, which does not vanish since the time-reversal symmetry is broken by the external magnetic field [9–11]. Here, we define the symmetric (+) and antisymmetric (−) parts of the tensor, $\mathcal{T}$, as

$$\mathcal{T}^{\pm}_{\mu\nu} := (\mathcal{T}_{\mu\nu} \pm \mathcal{T}_{\nu\mu})/2. \tag{1}$$

Notice that one can imagine various types of time correlations in quantum systems that approach $\langle j_\nu(0) j_\mu(t)\rangle_{\rm eq}$ in the classical limit, where $\langle\bullet\rangle_{\rm eq}$ denotes the equilibrium expectation value. Therefore, we first need to choose which type of time correlation should be employed in the FDT.

The answer was found in the measurement of the time correlation. Since we first measure $\hat{j}_\nu$ and then $\hat{j}_\mu$ after a certain time interval t, the measurement of $\hat{j}_\mu$ is disturbed by the preceding measurement of $\hat{j}_\nu$ in quantum systems. The uncertainty relation between measurement error and disturbance [39–41] does not allow this disturbance to vanish, since the measurement error should be smaller than the current fluctuation in the equilibrium. References [9,10] showed that such a disturbance effect can be minimized by adopting the quasiclassical measurement, which simulates the ideal classical measurement as closely as possible (see the references for its precise definition). They proved that the time correlation, taking into account such disturbance by the measurement, is symmetrized as

$$\Xi_{\mu\nu}(t) = \left\langle \tfrac{1}{2}\{\hat{j}_\nu(0), \hat{j}_\mu(t)\}\right\rangle_{\rm eq}, \tag{2}$$

where $\{A, B\} := AB + BA$. It is noteworthy that the disturbance by the measurement on the time correlation is relevant even in the macroscopic limit, $\Omega \to +\infty$. Actually, the disturbance on $\hat{j}_\mu$ caused by the preceding quasiclassical measurement of $\hat{j}_\nu$ was shown to be $O(1)$ [9,10].

In contrast, the definition of $\sigma_{\mu\nu}$ is essentially free from the disturbance caused by the measurement. Indeed, as was already pointed out in Kubo's original paper [6], observation of $\sigma_{\mu\nu}$ does not require the sequential measurements. One first prepares an ensemble of systems in the equilibrium and then applies a weak electric field along the ν axis. In each system, the induced current density $\hat{J}_\mu/\Omega$ is measured at a certain time t only once. By choosing different t for the measurement in each system, one can obtain the induced current as a function of t, no matter how large the disturbance by the measurement is.

In usual experiments, however, measurements are made sequentially at various t in a single system. Still, one can avoid the disturbance effect by adopting the quasiclassical measurement. Actually, the disturbance on $\hat{J}_\mu/\Omega = \hat{j}_\mu/\sqrt{\Omega}$ caused by a preceding quasiclassical measurement of J_μ/Ω is $O(1/\sqrt{\Omega})$, which is negligible in the macroscopic limit, $\Omega \to \infty$.

Consequently, the observed $\sigma_{\mu\nu}$ is given by the Kubo formula,

$$\sigma_{\mu\nu} = \theta(t)\beta\langle \hat{j}_\nu(0); \hat{j}_\mu(t)\rangle, \tag{3}$$

which was derived without considering the disturbance effect [6]. Here, the step function $\theta(t)$ represents the causality, and

$$\langle \hat{j}_\nu(0); \hat{j}_\mu(t)\rangle := \frac{1}{\beta}\int_0^\beta \langle e^{\lambda \hat{H}_{\rm eq}} \hat{j}_\nu(0) e^{-\lambda \hat{H}_{\rm eq}} \hat{j}_\mu(t)\rangle_{\rm eq} d\lambda \tag{4}$$

is called the canonical time correlation where $\hat{H}_{\rm eq}$ is the equilibrium Hamiltonian. Since it is different from the symmetrized time correlation of Eq. (2), the FDT, as a relation between the observed $\Xi_{\mu\nu}$ and $\sigma_{\mu\nu}$, can be violated.

If the types of measurements other than the quasiclassical ones are applied to the observation of the time correlation, the FDT violation would be further enhanced. Such enhancement is out of scope of the present study. We focus only on the FDT violation caused by the intrinsic reason.

In Ref. [6], Kubo already compared the conductivity,

$$\sigma_{\mu\nu}(\omega) = \int_0^\infty \beta\langle \hat{j}_\nu(0); \hat{j}_\mu(t)\rangle e^{i\omega t} dt,$$

with the spectral intensity,

$$\tilde{S}_{\mu\nu}(\omega) := \int_{-\infty}^{\infty} \Xi_{\mu\nu}(t) e^{i\omega t} dt, \tag{5}$$

without examining the measurability of the symmetrized time correlation, $\Xi_{\mu\nu}(t)$. He derived the identities,

$$\beta {\rm Re}\tilde{S}_{xx}(\omega) = 2I_\beta(\omega){\rm Re}\sigma_{xx}(\omega), \tag{6}$$

$$\beta {\rm Im}\tilde{S}_{xy}(\omega) = 2I_\beta(\omega){\rm Im}\sigma_{xy}(\omega), \tag{7}$$

with

$$I_\beta(\omega) := \frac{\beta\hbar\omega}{2}\coth\frac{\beta\hbar\omega}{2} \sim \begin{cases} 1 & (\beta\hbar|\omega| \ll 1), \\ \beta\hbar|\omega|/2 & (\beta\hbar|\omega| \gg 1). \end{cases} \tag{8}$$

References [9,10] not only proved the measurability of $\Xi_{\mu\nu}(t)$ but also showed that the spectral intensity should be redefined as

$$S_{\mu\nu}(\omega) := \int_0^\infty \Xi_{\mu\nu}(t)e^{i\omega t}dt, \tag{9}$$

which differs from $\tilde{S}_{\mu\nu}(\omega)$ in the lower bound of the integral. Indeed, the FDT for the Hall conductivity would be violated, $\sigma_{xy} \neq \beta\tilde{S}_{xy}$, if $\tilde{S}_{xy}$ were employed as 'fluctuation', even in classical systems [10]. To avoid such superficial violation caused by the improper choice of fluctuation in the frequency domain, we employ $S_{\mu\nu}$ as the proper definition of fluctuation.

We now focus on the violation in the DC limit, $\omega \to 0$, so that $\hbar\omega \ll T$ at any nonvanishing temperature. To quantify the violation, we rewrite Eqs. (C.3) and (C.5) in Ref. [10] as

$$\beta S_{xx}(0) = \beta \mathrm{Re} S_{xx}(0) = \frac{1}{2}\beta \mathrm{Re}\tilde{S}_{xx}(0) \tag{10}$$

$$\beta S_{xy}(0) = \beta \mathrm{Re} S_{xy}(0) = \int_{-\infty}^{\infty} \frac{\mathcal{P}}{\omega}\beta \mathrm{Im}\tilde{S}_{xy}(\omega)\frac{d\omega}{2\pi}. \tag{11}$$

Then, we obtain

$$\beta S_{xx}(0) = \sigma_{xx}(0), \tag{12}$$

$$\beta S_{xy}(0) = \int_{-\infty}^{\infty} \frac{\mathcal{P}}{\omega} I_\beta(\omega)\mathrm{Im}\sigma_{xy}(\omega)\frac{d\omega}{\pi}, \tag{13}$$

from Eqs. (6) and (7), $I_\beta(0) = 1$ and $\sigma_{xx}(0) = \mathrm{Re}\sigma_{xx}(0)$, where $\mathcal{P}$ denotes the principal value.

The FDT for the diagonal conductivity, $\beta S_{xx}(0) = \sigma_{xx}(0)$, is valid in both classical and quantum systems, supporting the naive Nyquist's argument [3]. In contrast, the FDT for the Hall conductivity, $\beta S_{xy}(0) = \sigma_{xy}(0)$, holds only in the classical systems. Actually, the dispersion relation [11],

$$\sigma_{xy}(0) = \mathrm{Re}\sigma_{xy}(0) = \int_{-\infty}^{\infty} \frac{\mathcal{P}}{\omega}\mathrm{Im}\sigma_{xy}(\omega)\frac{d\omega}{\pi} \tag{14}$$

shows that $\beta S_{xy}(0)$, given as Eq. (13), does differ from $\sigma_{xy}(0)$ by the extra factor, $I_\beta(\omega)$, in the integrand. This factor describes a genuine quantum effect, since $I_\beta(\omega) = 1$ in the classical limit, $\hbar \to 0$.

Our question is how large the quantum FDT violation, $|\sigma_{xy}(0) - \beta S_{xy}(0)|$, should be. We will demonstrate that it can overwhelm the typical value of $|\sigma_{xy}(0)|$ and even diverge in the low temperature limit.

III. CONDITIONS FOR LARGER VIOLATION

Equations (13) and (14) show that the FDT would not be violated if $\mathrm{Im}\sigma_{xy}(\omega)$ had nonzero values only at $\hbar\omega \ll T$. On the other hand, the optical absorption (cyclotron resonance) spectra for the left (L) and right (R) circularly polarized light are proportional to the real part of [73]

$$\sigma_{xy}^{\mathrm{L/R}}(\omega) := \sigma_{xx}(\omega) \pm i\sigma_{xy}(\omega). \tag{15}$$

Hence, $\mathrm{Im}\sigma_{xy}(\omega)$ should have positive and negative peaks at around $\omega = -\omega_\mathrm{c}$ and $+\omega_\mathrm{c}$, respectively, where $\omega_\mathrm{c} := eB/m$ is the cyclotron frequency. This is shown in Fig. 1(a), which is obtained by the method that we shall explain shortly. Therefore, we can expect significant FDT violation when

$$T \lesssim \hbar\omega_\mathrm{c}. \tag{16}$$

As we will show in Sec. VII, the violation is further enhanced, if the spectral peak at around $\omega = \pm\omega_\mathrm{c}$ is narrow,

$$\hbar\omega_\mathrm{c} \gtrsim 2\Gamma, \tag{17}$$

where Γ denotes the half width of the Landau level which is caused by the impurity scattering [57,58]. We henceforth assume inequalities (16) and (17) for T and B. The integer QHE can also be expected at sufficiently low temperature, $T \ll \hbar\omega_\mathrm{c} - 2\Gamma$, whereas such extremely low temperature (or QHE) is not necessarily required for observing the FDT violation clearly.

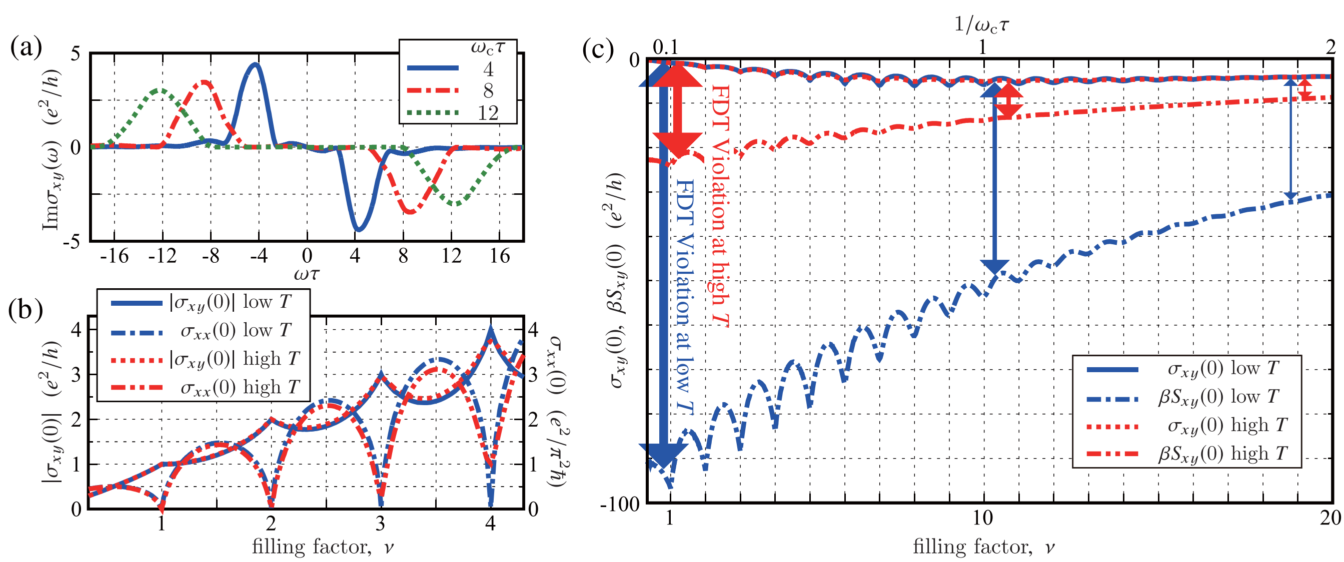

FIG. 1. (a) $\mathrm{Im}\sigma_{xy}(\omega)$ as a function of frequency ω for $\omega_\mathrm{c}\tau = 4$, 8, and 12 at temperature, $T = \hbar/20\tau$, and electron density, $n = 10m/h\tau$. Cyclotron frequency and the scattering time at $B = 0$ are denoted as ω_c and τ, respectively. (b) $|\sigma_{xy}(0)|$ and $\sigma_{xx}(0)$ as a function of filling factor, ν, at $T = \hbar/20\tau$ (low T) and $T = \hbar/5\tau$ (high T) for fixed electron density, $n = 10m/h\tau$. (c) $\sigma_{xy}(0)$ and $\beta S_{xy}(0)$ as a function of ν for the same values of parameters as in (b).

IV. MODEL AND METHOD OF CALCULATION

To avoid the inconsistency, which could arise due to the separate evaluation at different levels of approximation, we evaluate not only the FDT violation, $\sigma_{xy}(0) - \beta S_{xy}(0)$, but also each of $\beta S_{xy}(0)$ and $\sigma_{xy}(0)$ from a single quantity, $\mathrm{Im}\sigma_{xy}(\omega)$, by means of Eqs. (13) and (14).

To calculate $\mathrm{Im}\sigma_{xy}(\omega)$, we assume noninteracting electrons (with a charge $-e$, mass m) in two dimensions with the following single-body Hamiltonian:

$$\hat{H} = \frac{1}{2m}(\hat{\boldsymbol{p}} + e\boldsymbol{A}(\hat{\boldsymbol{r}}))^2 + \sum_i V_0 \delta(\hat{\boldsymbol{r}} - \boldsymbol{R}_i). \tag{18}$$

Here, $\boldsymbol{A} = (0, Bx)$ is the vector potential, $\boldsymbol{R}_i$ the location (obeying the uniform distribution) of an impurity, and V_0 is the strength of the impurity potential. The coordinate and momentum operators of the electron are denoted as $\hat{\boldsymbol{r}}$ and $\hat{\boldsymbol{p}}$, respectively.

To avoid artificial divergences caused by subtle treatment of the Landau level degeneracies, we employ the self-consistent Born approximation (SCBA) [57,58]. It properly gives the quantized Hall conductivity $\sigma_{xy}(0) = -\nu e^2/h$, when the filling factor $\nu := 2\pi l^2 n = nh/eB$ is an integer. Here, $l = \sqrt{\hbar/eB}$ is the magnetic length and n the electron density. The formula derived in Ref. [74], which is useful to calculate $\sigma_{xy}(0)$ [57,58], is inconvenient for our purpose, i.e., to evaluate $\mathrm{Im}\sigma_{xy}(\omega)$ for all ω including $\omega \sim \pm\omega_c$. We thus perform a straightforward calculation of the Kubo formula using the Landau-level basis and the SCBA. Then, we can obtain $\mathrm{Im}\sigma_{xy}(\omega)$ by solving the following self-consistent equation for the self-energy $\Sigma(\epsilon)$,

$$\Sigma(\epsilon) = \frac{n_i V_0^2}{2\pi l^2} \sum_N \frac{1}{\epsilon - \hbar\omega_c(N + 1/2) - \Sigma(\epsilon)}, \tag{19}$$

where N is the Landau index and n_i is the density of impurities. We can also estimate the half width of the Landau level Γ from $\Sigma(\epsilon)$. It has the asymptotic form [57,58],

$$\Gamma \sim \sqrt{\frac{2}{\pi}\frac{\hbar}{\tau}\hbar\omega_c}, \quad (\omega_c\tau \to \infty), \tag{20}$$

where the scattering time at $B = 0$ is denoted as

$$\tau = \left[\frac{2\pi}{\hbar} n_i V_0^2 \frac{m}{2\pi\hbar^2}\right]^{-1}. \tag{21}$$

V. DIAGONAL AND HALL CONDUCTIVITIES

We first confirm that the quantities evaluated from $\mathrm{Im}\sigma_{xy}(\omega)$ agree with the ones in the previous works calculated directly without referring to $\mathrm{Im}\sigma_{xy}(\omega)$ [57]. Figure 1(b) shows $|\sigma_{xy}(0)|$ $(= -\sigma_{xy}(0))$, obtained from Eq. (14), as a function of the filling factor ν at $T = \hbar/20\tau$ and $\hbar/5\tau$. The electron density is fixed to $n = 10m/h\tau$, and the increase of ν implies the decrease of B. Inequalities (16) and (17) hold at $\nu \lesssim 4$. At low temperature, $T = \hbar/20\tau$, integer QHE is expected. Indeed, $\sigma_{xy}(0)$ is quantized to $-\nu e^2/h$ at $\nu = 1$, 2, 3, and 4, in agreement with the previous theories [57] and experiments [59–61]. At high temperature, $T = \hbar/5\tau$, the quantization blurs at $\nu = 3$ and 4, because thermal excitation to the higher Landau levels takes place. Since these results are obtained by integrating $\mathrm{Im}\sigma_{xy}(\omega)$, they indicate that $\mathrm{Im}\sigma_{xy}(\omega)$ is reasonably obtained for all ω.

We also calculate the diagonal conductivity $\sigma_{xx}(0)$ as shown in Fig. 1(b). It is nearly quantized as $\sigma_{xx}(0) = (e^2/\pi^2\hbar)\nu$ for half-integer values of ν, in agreement with the previous studies [58,60–62]. When an external electric field is applied in the x direction, dissipation occurs if $\sigma_{xx} > 0$. This happens for every ν, except when the QHE takes place at integer ν and at low T.

VI. RESULTS FOR FDT VIOLATION

In Fig. 1(c), the difference between $\sigma_{xy}(0)$ and $\beta S_{xy}(0)$ indicates the magnitude of violation of FDT as a function of ν. Here, the scale of the vertical axis is about 20 times larger than that of Fig. 1(b).

The magnitude of the violation is larger than $|\sigma_{xy}(0)|$ at $T = \hbar/5\tau$ ($\sim$1 K for $\tau \sim 10^{-12}$ s). It is further enhanced, at low temperature $T = \hbar/20\tau$, to an order of magnitude larger than $|\sigma_{xy}(0)|$. This counterintuitive result should be contrasted with the naive conjectures mentioned in Sec. I.

The violation is enhanced with decreasing ν and lowering T. This result confirms the expectation in Sec. III that the FDT is violated significantly when $\hbar\omega_c \gtrsim T$.

VII. GENERAL CONSIDERATION

Let us extend our argument to the general macroscopic systems (such as ones with many-body interactions) and to the LRs of the general macroscopic observable of a 'current' vector $\dot{\hat{\boldsymbol{a}}} := d\hat{\boldsymbol{a}}/dt$, associated with the 'displacement' vector $\hat{\boldsymbol{a}}$. Following Ref. [10], we compare the 'fluctuation' between two components of $\Delta\dot{\hat{\boldsymbol{a}}} := \dot{\hat{\boldsymbol{a}}} - \langle\dot{\hat{\boldsymbol{a}}}\rangle_{\mathrm{eq}}$,

$$S_{\mu\nu}(\omega) := \int_0^\infty \left\langle \frac{1}{2}\{\Delta\dot{\hat{a}}_\nu(0), \Delta\dot{\hat{a}}_\mu(t)\}\right\rangle_{\mathrm{eq}} e^{i\omega t} dt, \tag{22}$$

with the admittance tensor, $\chi_{\mu\nu}(\omega)$, describing the LR of $\dot{\hat{a}}_\mu$ to the external field coupled to $\hat{a}_\nu$. When the symmetric and antisymmetric parts are defined as Eq. (1), the power loss or the energy dissipation of the external field of frequency ω is determined only by $\mathrm{Re}\chi^+_{\mu\nu}(\omega)$ and $\mathrm{Im}\chi^-_{\mu\nu}(\omega)$, but is independent of $\mathrm{Im}\chi^+_{\mu\nu}(\omega)$ and $\mathrm{Re}\chi^-_{\mu\nu}(\omega)$ [11].

In classical systems, the FDT, $\chi_{\mu\nu}(\omega) = \beta S_{\mu\nu}(\omega)$, rigorously holds for any choice of μ, ν, and ω [8–10]. However, in the quantum systems, it could be violated regarding the dissipationless component even at $\omega = 0$ as $\chi^-_{\mu\nu}(0) \neq \beta S^-_{\mu\nu}(0)$, whereas the dissipative component still obeys the FDT as $\chi^+_{\mu\nu}(0) = \beta S^+_{\mu\nu}(0)$ [9,10]. [Note that $\chi^\pm_{\mu\nu}(0)$ and $S^\pm_{\mu\nu}(0)$ are real.] Actually, one can straightforwardly generalize Eq. (13) to

$$\beta S^-_{\mu\nu}(0) = \int_{-\infty}^\infty \frac{\mathcal{P}}{\omega} I_\beta(\omega) \mathrm{Im}\chi^-_{\mu\nu}(\omega) \frac{d\omega}{\pi}, \tag{23}$$

whereas the dispersion relation [11] gives

$$\chi^-_{\mu\nu}(0) = \int_{-\infty}^\infty \frac{\mathcal{P}}{\omega} \mathrm{Im}\chi^-_{\mu\nu}(\omega) \frac{d\omega}{\pi}. \tag{24}$$

Again, the FDT is violated because of the extra factor $I_\beta(\omega)$ in Eq. (23). Note that $\chi^-_{\mu\nu} = S^-_{\mu\nu} = 0$ if the system has the time-reversal symmetry [10]. Hence, the violation occurs only when the time-reversal symmetry is broken.

To estimate the magnitude of the violation,

$$\chi^-_{\mu\nu}(0)-\beta S^-_{\mu\nu}(0) = \int_{-\infty}^{\infty} \mathcal{P}\frac{1-I_\beta(\omega)}{\omega}\mathrm{Im}\chi^-_{\mu\nu}(\omega)\frac{d\omega}{\pi}, \quad (25)$$

we rewrite it in terms of

$$\chi^{\mathrm{L}}_{\mu\nu}(\omega) := [\chi_{\mu\mu}(\omega)+\chi_{\nu\nu}(\omega)]/2 + i\chi^-_{\mu\nu}(\omega), \quad (26)$$

as

$$\chi^-_{\mu\nu}(0)-\beta S^-_{\mu\nu}(0) = \beta\int_{-\infty}^{\infty} \mathcal{P}\frac{I_\beta(\omega)-1}{\beta\omega}\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)\frac{d\omega}{\pi}. \quad (27)$$

Since $\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)$, which corresponds to the real part of (15), describes the power absorption spectrum of a rotating external field, it should be nonnegative according to the second law. Moreover, $[I_\beta(\omega)-1]/\beta\omega$ vanishes at $\beta\hbar|\omega| \ll 1$, whereas it can be approximated to the sign function, $\mathrm{sgn}(\omega)$, at $\beta\hbar|\omega| \gg 1$. Hence, the magnitude of the FDT violation is roughly determined by the difference between the spectral intensities of $\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)$ distributed in $\beta\hbar\omega \gtrsim 1$ and in $\beta\hbar\omega \lesssim -1$. Thus, we can expect significant violation at

$$T \lesssim \hbar\bar{\omega} := \hbar\frac{\left|\int_{-\infty}^{\infty}\omega\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)d\omega/\pi\right|}{\int_{-\infty}^{\infty}\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)d\omega/\pi}, \quad (28)$$

as long as the spectral first moment $\bar{\omega}$ is finite. Note that $\bar{\omega}$ vanishes for systems with time-reversal symmetry, where $\mathrm{Im}\chi^-_{\mu\nu}(\omega)$, the odd component of $\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)$, vanishes. To evaluate $\bar{\omega}$, we can use the moment sum rule [11],

$$\int_{-\infty}^{\infty}\omega^n\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)\frac{d\omega}{\pi} = \frac{i(-i)^n}{\hbar}\left\langle\left[\dot{\hat{c}}_-, \frac{d^n\hat{c}_+}{dt^n}\right]\right\rangle_{\mathrm{eq}}, \quad (29)$$

with $n=0,1$ and $\hat{c}_\pm := (\hat{a}_\mu \pm i\hat{a}_\nu)/\sqrt{2}$. The violation even diverges in proportion to β at $T\to 0$ with the asymptotic form,

$$\chi^-_{\mu\nu}(0)-\beta S^-_{\mu\nu}(0) \sim \beta\int_{-\infty}^{\infty}\mathrm{sgn}(\omega)\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)\frac{d\omega}{\pi}, \quad (30)$$

if the integral in the rhs is not canceled out.

As for our example of the Hall conductivity, $\hat{\boldsymbol{a}}$ stands for $-e\sum_i \hat{\boldsymbol{r}}_i/\sqrt{\Omega}$, where $\hat{\boldsymbol{r}}_i$ is the coordinate operator of each electron. Thus, inequality (28) reproduces (16), since Eq. (29) is explicitly calculated, for $n=0$ and 1, as

$$\int_{-\infty}^{\infty}\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)\frac{d\omega}{\pi} = \frac{ne^2}{m}, \quad (31)$$

$$\int_{-\infty}^{\infty}\omega\mathrm{Re}\chi^{\mathrm{L}}_{\mu\nu}(\omega)\frac{d\omega}{\pi} = \frac{ne^3B}{m^2}. \quad (32)$$

The rhs of Eq. (30) is further enhanced at larger $\hbar\omega_c/2\Gamma$, since the spectral intensity of $\mathrm{Re}\sigma^{\mathrm{L}}_{xy}(\omega)$ is more localized at $\omega > 0$. This yields the condition (17). In the limit of $\omega_c\tau\to\infty$ and $T\to 0$, we have $\hbar\omega_c/2\Gamma\to\infty$ and $|\sigma_{xy}(0)| \sim \nu e^2/h = ne/B$, and

$$\frac{|\sigma_{xy}(0)-\beta S_{xy}(0)|}{|\sigma_{xy}(0)|} \sim \frac{\beta\hbar\omega_c}{2}. \quad (33)$$

For $n = 10m/h\tau$, $\nu = 1$ and $T = \hbar/20\tau$, the rhs of Eq. (33) equals to 10^2, which is consistent with our data shown in Fig. 1(c).

VIII. ESTIMATION OF FLUCTUATION FROM LR FUNCTION

The FDT has been widely utilized to estimate the fluctuations (noises) from the LRs when designing, e.g., electric circuits [31–33], optical devices [33–35], and gravitational-wave detectors [36]. However, we should be careful in the quantum system where the time-reversal symmetry is broken, because the FDT can severely underestimate the fluctuations, if it is applied to the dissipationless components of LRs. This is true even at $\hbar\omega \ll T$, where, by contrast, the FDT still exactly holds for the dissipative components of LRs. Therefore, alternative formulas that are correct in this case are craved. Equations (13) and (23) are such formulas, by which one can estimate fluctuations from the observed LR functions for dissipationless components. They will be helpful in practical applications.

IX. NOTES

Although Fig. 1 is obtained approximately for a noninteracting system with short-range impurities, our general argument based on the sum rules is rigorous and independent of the details of the impurities and the electron-electron interaction. Therefore, the FDT violation in the Hall conductivity should be *universally* observed in 2DES in a magnetic field, as long as inequalities (16) and (17) are fulfilled. For example, in a Si inversion layer sample [60] with $m \sim 10^{-1}m_0$ (m_0: free electron mass), $n \sim 10^{11}$ cm^{-2}, and the mobility $\mu = e\tau/m = \omega_c\tau/B \sim 10^4$ cm^2/Vs, should show relevant FDT violation in a high magnetic field $B \gtrsim 2$ T and at low temperature $T \lesssim 20$ K.

The caveat is that S_{xy}, rather than $\tilde{S}_{xy}$, should be compared with σ_{xy}, to let the FDT (in the frequency domain) valid in the classical systems [10]. If the spectrum analyzer yields $\tilde{S}_{xy}$, one needs to convert it to S_{xy} using Eqs. (C.3)–(C.6) in Ref. [10].

X. SUMMARY

We have studied the FDT as a relation between the observed LR functions and the observed fluctuations in macroscopic systems, assuming that the measurements are as ideal as possible and that the system is close to equilibrium. It is found that the FDT can be violated even at $\omega = 0$ by a macroscopically large magnitude regarding the dissipationless components of LRs of 'current.' We have also proposed an alternative method which can evaluate the fluctuations from the LRs precisely even in such a case. These results are indispensable for the understanding of the nonequilibrium physics and for practical applications.

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