Time-Asymmetric Fluctuations of Light and the Breakdown of Detailed Balance

A. Denisov,[1] H. M. Castro-Beltran,[2] and H. J. Carmichael[1]

[1]Department of Physics, University of Oregon, Eugene, Oregon 97403-1274

[2]Centro de Investigacion en Ingenieria y Ciencias Aplicadas, Universidad Autonoma del Estado de Morelos, Avenida Universidad 1001, 62210 Cuernavaca, Morelos, Mexico

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Temporal fluctuations of the light radiated by a photoemissive source are studied through the cross correlation of output fields. Whereas microscopic reversibility guarantees time-symmetric fluctuations in thermal equilibrium—where detailed balance holds—away from equilibrium time asymmetry is permitted. Examples of time asymmetry in cavity QED are reported.

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The time symmetry of the cross correlation of fluctuations about equilibrium, $\langle B(t+\tau)A(t)\rangle = \langle B(t-\tau)A(t)\rangle$, where A and B are state variables, has a central place in statistical physics; it provides the fundamental basis for the Onsager relations [1,2]. The symmetry follows from microscopic reversibility (A and B are assumed symmetric under time reversal), which requires that the equilibrium be maintained in detailed balance [3]. In quantum optics, one is generally concerned with steady states away from equilibrium, where correlation functions of the light emitted by an open system (photoemissive source) are measured through photoelectric detection. The detected radiation field is an outgoing field that is absorbed by the environment, and its steady state is manifestly not symmetric under time reversal. Fluctuations about the steady state may therefore exhibit a specific time order.

Studies of fluctuations in quantum optics have focused, nonetheless, on time-symmetric correlations. One reason is that the nonclassical phenomena of photon antibunching and squeezing deal with autocorrelations, $\langle A(t+\tau)A(t)\rangle$, which are symmetric by definition for a stationary process. Another is that, although detailed balance is not guaranteed to hold away from equilibrium [4,5], it may still do so due to boundary conditions and symmetry [6]; it was noted in the early 1970s, for example, that the laser satisfies detailed balance [7], in spite of the fact that it operates far from thermal equilibrium. A few quantum optics papers have addressed issues related to time-ordered transitions in resonance fluorescence [8–10] and quantum dots [11]; none, however, deal with the breakdown of detailed balance.

Detailed balance is traditionally defined as the balancing of transitions in equilibrium between pairs of quantum states [4], or, for a Markov process on a "classical" phase space, the vanishing of the equilibrium probability current [5]. The general situation in quantum optics cannot be approached in the first of these ways, since the presence of coherence disallows the assumed perturbative dynamics. The second, however, may be stated explicitly as a condition on the two-time quasiprobability density in the P representation,

$$P(\boldsymbol{x}, t+\tau; \boldsymbol{x}_0, t) = P(\tilde{\boldsymbol{x}}, t-\tau; \tilde{\boldsymbol{x}}_0, t), \qquad (1)$$

where $\boldsymbol{x}$ and $\boldsymbol{x}_0$ are any two phase-space states, and the tilde (˜) denotes time reversal. This phase-space approach holds in a strict sense only when the dynamics in the P representation is classical—i.e., when the quasidistribution is nonsingular and positive definite. We may adopt it, by extrapolation, though, also when the fluctuations are "nonclassical" (P is a highly singular distribution) and there is no strict phase-space probability current. Condition (1) then carries over as a statement of time symmetry for two-time operator averages [12,13], the operators placed in time and normal order. We thus define detailed balance in an operational sense which holds for measurements made through photodetection on outgoing fields.

Graham and Haken [7] accounted for the parallels between the laser and an equilibrium system undergoing a second-order phase transition by observing that the laser Fokker-Planck equation satisfies the conditions for detailed balance. Tomita and Tomita [5], noting that detailed balance is not expected to hold away from equilibrium, and considering the case of Gaussian fluctuations, stated what is required in addition to a nonequilibrium flux through the system if detailed balance is to fail: "(The) existence of a coupling between more than one degree of freedom, so that there can be a *direction* in the through flux." Such a coupling—between the atom(s) and the cavity field—is the central feature of cavity QED. In this Letter, we report results for correlation functions in cavity QED that exhibit time asymmetries demonstrating the breakdown of detailed balance. Two specific measurement scenarios are considered (Fig. 1): the cross correlation of intensities measured in distinct output channels, and the correlation in a single channel of intensity and field amplitude through condi-

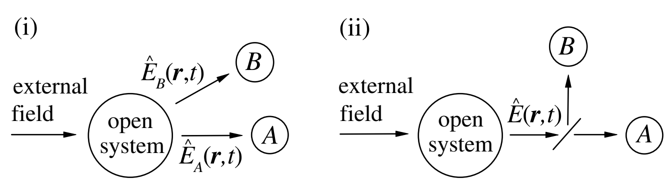

FIG. 1. Two setups for the cross correlation of output fields. A trigger photodetection at A is correlated with (i) a photodetection at B and (ii) a field amplitude measurement at B.

tional homodyne detection [14,15]. We demonstrate time asymmetry of the intensity-intensity cross correlation for arbitrarily weak excitation and for Gaussian fluctuations, while conditional homodyne detection yields asymmetry only within an intermediate range of excitation where the fluctuations are non-Gaussian.

We begin by demonstrating the breakdown of detailed balance for coupled classical harmonic oscillators. Consider two oscillators, A and B, interacting with reservoirs at different temperatures having thermal photon numbers $\bar{n}_A$ and $\bar{n}_B$, respectively. The oscillators are coupled in the rotating-wave approximation with coupling constant g, such that their amplitudes, α and β, obey the Langevin equations,

$$d\alpha = (-\kappa_A \alpha + g\beta)dt + \sqrt{2\kappa_A \bar{n}_A}\, dZ_A\,, \qquad (2a)$$

$$d\beta = (-\kappa_B \beta - g\alpha)dt + \sqrt{2\kappa_B \bar{n}_B}\, dZ_B\,, \qquad (2b)$$

where κ_A and κ_B are damping rates, and dZ_A and dZ_B are independent complex-valued Wiener increments; $\langle dZ_\mu dZ_\mu \rangle = 0$, $\langle dZ_\mu^* dZ_\mu \rangle = dt$. The Langevin equations yield coupled equations for the sets of correlation functions (regression formula),

$$G_{AA}^{(2)}(\tau) \equiv \langle |\alpha(t+\tau)|^2 |\alpha(t)|^2 \rangle\,, \qquad (3a)$$

$$G_{AB}^{(2)}(\tau) \equiv \langle |\beta(t+\tau)|^2 |\alpha(t)|^2 \rangle\,, \qquad (3b)$$

$$G_{AC}^{(2)}(\tau) \equiv \mathrm{Re}\langle (\alpha\beta^*)(t+\tau)\, |\alpha(t)|^2 \rangle\,, \qquad \tau \geq 0\,, \qquad (3c)$$

and

$$G_{AB}^{(2)}(\tau) \equiv \langle |\alpha(t-\tau)|^2 |\beta(t)|^2 \rangle\,, \qquad (4a)$$

$$G_{BB}^{(2)}(\tau) \equiv \langle |\beta(t-\tau)|^2 |\beta(t)|^2 \rangle\,, \qquad (4b)$$

$$G_{CB}^{(2)}(\tau) \equiv \mathrm{Re}\langle (\alpha\beta^*)(t-\tau)\, |\beta(t)|^2 \rangle, \qquad \tau \leq 0\,. \qquad (4c)$$

When the reservoir temperatures are equal ($\bar{n}_A = \bar{n}_B$), the intensity cross correlation $G_{AB}^{(2)}$ is found to be time symmetric. The symmetry fails, however, if $n_A \neq n_B$, as illustrated by the examples in Fig. 2 (normalized correlation functions are plotted in all figures).

The classical oscillator example maps to an example in quantum optics by considering α and β to be the amplitudes in the P representation of quantized field modes. Denoting the mode creation and annihilation operators by $\hat{a}^\dagger$, $\hat{b}^\dagger$ and $\hat{a}$, $\hat{b}$, the intensity cross correlation displayed in Fig. 2 is the time-ordered, normal-ordered correlation function,

$$G_{AB}^{(2)}(\tau) = \begin{cases} \langle \hat{a}^\dagger(t)\hat{b}^\dagger(t+\tau)\hat{b}(t+\tau)\hat{a}(t)\rangle & \tau \geq 0 \\ \langle \hat{b}^\dagger(t)\hat{a}^\dagger(t-\tau)\hat{a}(t-\tau)\hat{b}(t)\rangle & \tau \leq 0\,. \end{cases} \qquad (5)$$

Making now the transcription to cavity QED, harmonic oscillator A is a mode of an optical cavity, while harmonic oscillator B is replaced by a two-state atom; thus, $\hat{b}^\dagger$ and $\hat{b}$ are replaced by atomic raising and lowering operators $\hat{\sigma}_+$ and $\hat{\sigma}_-$. The cavity mode is driven by an external field and the cross correlation of intensities is made by measuring the rate of coincidences between a photon transmitted by the cavity and a photon scattered by the atom—thus, the detection of side-scattered photons is either preselected ($\tau \geq 0$) or postselected ($\tau \leq 0$) by a forwards-scattered photodetection [Fig. 1(i)]. The measured correlation function is calculated as

$$G_{AB}^{(2)}(\tau) = \begin{cases} \mathrm{tr}\{\hat{\sigma}_+\hat{\sigma}_- e^{\mathcal{L}|\tau|}[\hat{a}\rho_{\mathrm{ss}}\hat{a}^\dagger]\} & \tau \geq 0 \\ \mathrm{tr}\{\hat{a}^\dagger\hat{a} e^{\mathcal{L}|\tau|}[\hat{\sigma}_-\rho_{\mathrm{ss}}\hat{\sigma}_+]\} & \tau \leq 0\,, \end{cases} \qquad (6)$$

where $\mathcal{L}\rho_{\mathrm{ss}} = 0$, and

$$\begin{aligned}\mathcal{L} \equiv \mathcal{L}_{\mathrm{ext}} &+ g[\hat{a}^\dagger\hat{\sigma}_- - \hat{a}\hat{\sigma}_+, \cdot] \\ &+ \kappa(2\hat{a}\cdot\hat{a}^\dagger - \hat{a}^\dagger\hat{a}\cdot - \cdot\hat{a}^\dagger\hat{a}) \\ &+ (\gamma/2)(2\hat{\sigma}_-\cdot\hat{\sigma}_+ - \hat{\sigma}_+\hat{\sigma}_-\cdot - \cdot\hat{\sigma}_+\hat{\sigma}_-) \end{aligned} \qquad (7)$$

is a superoperator, with

$$\begin{aligned}\mathcal{L}_{\mathrm{ext}} \equiv\; &\Gamma(2\hat{a}^\dagger\cdot\hat{a} - \hat{a}\hat{a}^\dagger\cdot - \cdot\hat{a}\hat{a}^\dagger) \\ &+ \Gamma(2\hat{a}\cdot\hat{a}^\dagger - \hat{a}^\dagger\hat{a}\cdot - \cdot\hat{a}^\dagger\hat{a})\,, \end{aligned} \qquad (8a)$$

$$\mathcal{L}_{\mathrm{ext}} \equiv \mathcal{E}[\hat{a}^\dagger - \hat{a}, \cdot] \qquad (8b)$$

for incoherent or coherent excitation of the cavity, respectively. The damping rates of the cavity field and atomic polarization are, respectively, κ and $\gamma/2$, and either Γ or $\mathcal{E}$ determines the strength of the excitation.

Figure 3 shows the cross correlation for weak incoherent excitation and all other parameters the same as in Fig. 2. As this figure reveals, for weak excitation, the similar physical systems show similar correlations. The regression of fluctuations is, in fact, the same in the two systems; $G_{AB}^{(2)}$ satisfies a set of equations—along with $G_{AA}^{(2)}$ and $G_{AC}^{(2)}(\tau \geq 0)$ or $G_{BB}^{(2)}$ and $G_{CB}^{(2)}(\tau \leq 0)$ —that are the same whether B is a harmonic oscillator or a two-state atom. The differences between Figs. 2(ii) and 3(ii) arise from the initial conditions, where for B a two-state atom, the identity $\hat{\sigma}_+^2 = \hat{\sigma}_-^2 = 0$ yields the distinctly quantum mechanical initial values, $G_{BB}^{(2)}(0_-) = G_{CB}^{(2)}(0_-) = 0$.

For coherent excitation, we follow the method in Ref. [16] to obtain a closed form expression for the normalized cross correlation in the weak-driving-field limit:

$$g_{AB}^{(2)}(\tau) = \left\{1 + \frac{2C_1\xi}{1+\xi+2C_1}\, e^{-(1/2)(\kappa+\gamma/2)|\tau|}\left[\cos(\Omega\tau) - \frac{(1+2C_1)\gamma - \mathrm{sgn}(\tau)(\kappa+\gamma/2)}{2\Omega}\sin(\Omega\tau)\right]\right\}^2, \qquad (9)$$

where $\Omega = [g^2 - \frac{1}{4}(\kappa - \gamma/2)^2]^{1/2}$ and $C_1 \equiv g^2/\kappa\gamma$, $\xi \equiv 2\kappa/\gamma$. The result is the square of a two-photon transition amplitude with the time asymmetry identified explicitly in the coefficient of $\sin(\Omega\tau)$. Figure 4(i) shows an example of an intensity cross correlation in agreement with Eq. (9). At higher excitation [Fig. 4(ii)] Eq. (9) does not hold, but the time asymmetry persists even into the strong excitation regime [Figs. 4(iii) and 4(iv)].

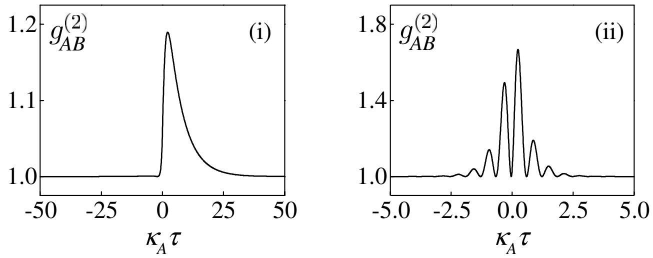

FIG. 2. Time asymmetry in the cross correlation of coupled harmonic oscillator intensities away from thermal equilibrium ($\bar{n}_A \neq 0$, $\bar{n}_B = 0$): (i) $g/\kappa_A = 0.1$, $\kappa_B/\kappa_A = 0.07$; (ii) $g/\kappa_A = 5.0$, $\kappa_B/\kappa_A = 1$.

In the second setup of Fig. 1(ii), the correlation function is measured through conditional homodyne detection of the light emitted into a single output channel. Although no direct cross correlation of coupled modes is made, the internal coupling is revealed through the cross correlation of different observables. The measured time- and normal-ordered correlation function is

$$2H(\tau) = \begin{cases} \langle \hat{a}^\dagger(t)\hat{a}(t+\tau)\hat{a}(t)\rangle + \text{c.c.} & \tau \geq 0 \\ \langle \hat{a}^\dagger(t-\tau)\hat{a}(t-\tau)\hat{a}(t)\rangle + \text{c.c.} & \tau \leq 0, \end{cases} \tag{10}$$

which involves an odd moment of the field amplitude; coherent excitation must therefore be used to ensure that the correlation function does not vanish.

This second scenario adds a new dimension. If the fluctuations are Gaussian about a nonzero mean field, $H(\tau)$ reduces to the autocorrelation of the field amplitude and is time symmetric by definition [14]. Thus, time asymmetry not only indicates the breakdown of detailed balance, it is also a direct probe of non-Gaussian fluctuations. Coupled harmonic oscillators have Gaussian fluctuations and cannot yield an asymmetric $H(\tau)$. We might look to resonance fluorescence, with its non-Gaussian fluctuations, for the simplest example of a time asymmetry. Resonant scattering involves transitions between *two* states only, though, and thus detailed balance is imposed by the low dimensionality [4]; indeed, $H(\tau)$ is found to be symmetric in resonance fluorescence.

It is not generally symmetric in cavity QED. We demonstrate time asymmetry for two cases, one where the fluctuations are nonclassical and the other where they are classical—a positive, nonsingular P distribution exists. We first consider intrinsic quantum fluctuations in a single-atom cavity QED system, where

$$2H(\tau) = \begin{cases} \mathrm{tr}[\hat{a}e^{\mathcal{L}|\tau|}(\hat{a}\rho_{\mathrm{ss}}\hat{a}^\dagger)] + \text{c.c.} & \tau \geq 0 \\ \mathrm{tr}[\hat{a}^\dagger\hat{a}e^{\mathcal{L}|\tau|}(\hat{a}\rho_{\mathrm{ss}})] + \text{c.c.} & \tau \leq 0, \end{cases} \tag{11}$$

with $\mathcal{L}$ defined in Eqs. (7) and (8). We then look to the related example of absorptive optical bistability with external noise, where the fluctuations take place in a classical phase space. Quantum fluctuations are made negligible by taking the number of atoms very large, and the noise source is taken, for simplicity, to be fluctuations of the external field amplitude $\mathcal{E}$. The correlation function is calculated as $\langle x^2(t)x(t+\tau)\rangle$, where $x(t)$ satisfies the stochastic Maxwell-Bloch equations,

$$dx = (-x + 2Cy + Y)(\kappa dt) + \Upsilon\sqrt{2\kappa}\, dW, \tag{12a}$$

$$dy = (-y + xz)(\gamma dt/2), \tag{12b}$$

$$dz = (-z - 1 - xy)(\gamma dt). \tag{12c}$$

$C \equiv NC_1$, $Y \equiv n_{\mathrm{sat}}^{-1/2}(\bar{\mathcal{E}}/\kappa)$, where $n_{\mathrm{sat}} = 8g^2/\gamma\kappa$ is the saturation photon number, $n_{\mathrm{sat}}x^2(t)$ is the intracavity photon number, and $y(t)/\sqrt{2}$ and $z(t)$ are the polarization and inversion per atom; Υ is the rms noise amplitude and dW is a Wiener increment.

Results are summarized in Fig. 5, on the left, for increasing strength of the external field in cavity QED [5(i)–5(iv)], and for increasing external noise in absorptive bistability on the right [5(v)–5(viii)]. The parameters have been chosen to give qualitatively similar results, not to correspond to the same operating conditions—-though decay rates and coupling strengths do match. In both the weak and the strong excitation limits, the correlation functions are time symmetric; time asymmetry is

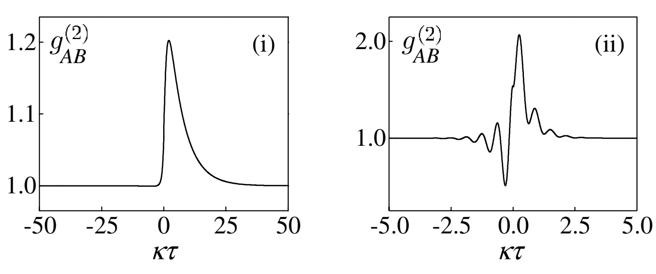

FIG. 3. Time asymmetry in the cross correlation of the side-scattered and forwards-scattered light intensities in cavity QED, with incoherent excitation: for a mean intracavity photon number $\langle \hat{a}^\dagger \hat{a}\rangle = 10^{-2}$ and (i) $g/\kappa = 0.1$, $\gamma/2\kappa = 0.07$; (ii) $g/\kappa = 5.0$, $\gamma/2\kappa = 1$.

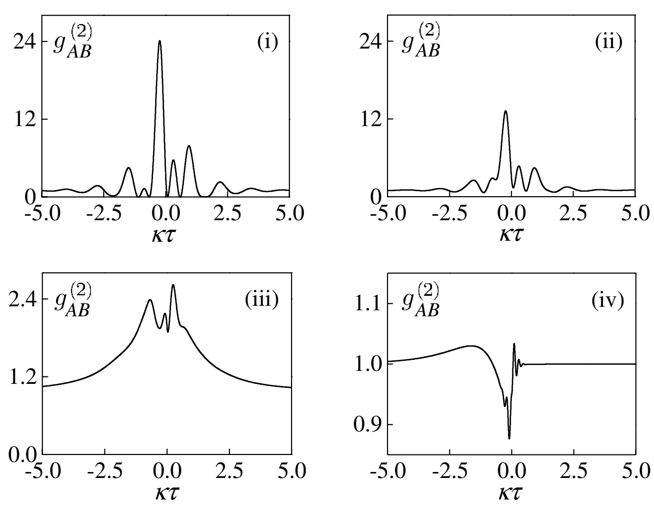

FIG. 4. Time asymmetry in the cross correlation of the side-scattered and forwards-scattered light intensities in cavity QED, with coherent excitation: for $g/\kappa = 5$, $\gamma/2\kappa = 1$, and mean intracavity photon numbers $\langle \hat{a}^\dagger \hat{a}\rangle = 10^{-5}$ (i), 10^{-3} (ii), 10^{-1} (iii), and 10 (iv).

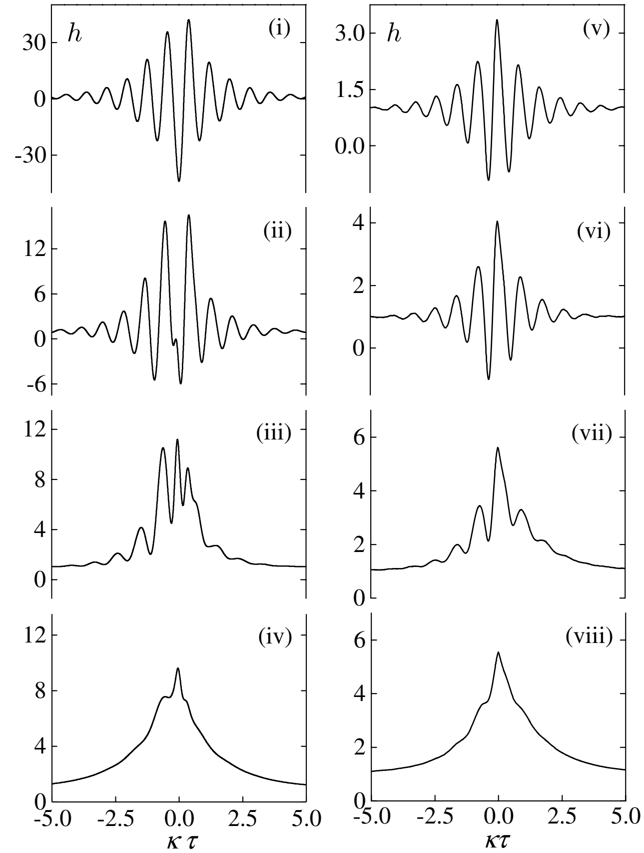

FIG. 5. Time asymmetry in the cross correlation of the intensity and field amplitude of the forwards-scattered light in cavity QED. Results for one atom and no external noise (left column) are compared with results for $N \gg 1$ atoms and amplitude noise on the external field (right column). All curves are for $\sqrt{N}\,g/\kappa = 8$ and $\gamma/\kappa = 1.25$. Those on the left are for intracavity photon numbers $\langle \hat{a}^\dagger \hat{a} \rangle = 10^{-4}$ (i), 10^{-3} (ii), 10^{-2} (iii), and 10^{-1} (iv); those on the right are for $Y = 13$ and $2Y^2 = 25$ (v), 50 (vi), 80 (vii), and 120 (viii).

restricted to a transition region in which the fluctuations are non-Gaussian.

Note how the oscillation is inverted and much larger in curves 5(i)–5(iv), compared with 5(v)–5(viii). These distinctly nonclassical features are discussed in [14]. An experiment in a many-atom cavity QED system (atomic beam) has observed the nonclassical inversion of the oscillation [15]. Although the number of atoms involved in the experiment is too large for us to make an exact calculation, we present, in Fig. 6, the results of an approximation which takes into account the five atoms coupled most strongly to the cavity mode; we increase the individual atom coupling strengths, accordingly, to recover the correct collective oscillation frequency. The main deficiency of this approximation is its overestimation of the dephasing caused by atomic beam fluctuations. In spite of this, time asymmetry is found in qualitative agreement with the experiment (Figs. 3(a) and 4(b) of Ref. [15]); in particular, we obtain the same change in the sign of the asymmetry for an increase in excitation strength.

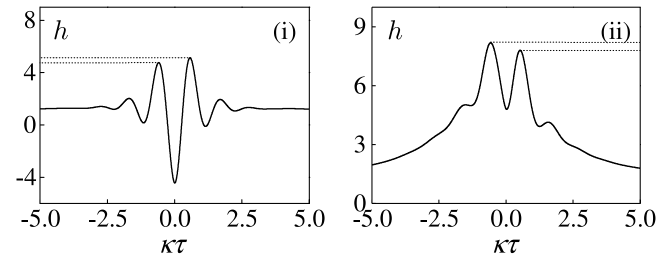

FIG. 6. As in Fig. 5, but for a many-atom cavity QED system without external noise. Results are averaged over 200 configurations of the five atoms most strongly coupled to a TEM_{00} standing-wave cavity mode $[g_j = g_0 \cos\theta_j \exp(-r_j^2/w_0^2)]$ assuming a uniform spatial distribution of atoms: for a density of $\bar{N}_{\rm eff} = 11$ atoms inside the mode waist $(r_j < w_0/2)$, $g_0/\kappa = 3.7$, $\gamma/\kappa = 1.25$, and mean intracavity photon numbers $\langle \hat{a}^\dagger \hat{a} \rangle \approx 2.1 \times 10^{-3}$ (i) and 7.3×10^{-3} (ii).

We have shown that fluctuations of the light emitted by cavity QED sources exhibit asymmetries in time associated with the breakdown of detailed balance. Conditional homodyne detection reveals time asymmetries that probe non-Gaussian fluctuations. Two-state scattering processes, such as resonance fluorescence, do not show these time asymmetries.

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