Violation of Leggett–Garg Inequalities in Single Quantum Dots*

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We investigate the violation of two types of Leggett–Garg (LG) inequalities in self-assembled quantum dots under the stationarity assumption. By comparing the two types of LG inequalities, we find the better one that is easier to be tested in an experiment. In addition, we show that the fine-structure splitting, background noise and temperature of quantum dots greatly affect the violation of LG inequalities.

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The transition from a strange quantum description to our familiar classical description is a fundamental problem in understanding the world. This issue was first investigated by Leggett and Garg in 1985[1] and led to the formulation of the so-called temporal Bell inequalities. Instead of considering the correlations between two spatially separated systems, they were concerned with the correlations of the state of a single system at different times. Furthermore, in order to obtain the inequalities, Leggett and Garg made two general assumptions:[2] (1) Any two-level macroscopic system will be at any time in one of the two accessible states (macroscopic realism). (2) The actual state of the system can be determined with arbitrarily small perturbation of its subsequent dynamics (non-invasive measurability). Leggett–Garg (LG) inequalities provide a criterion to characterize the boundary between the quantum realm and the classical realm and the possibility of identifying the macroscopic quantum coherence.

There have been many proposals to test such kind of inequalities by employing superconducting quantum interference devices.[3–5] However, owing to the extreme difficulty of experiments with truly macroscopic systems and the requirement for a noninvasive measurement which describes the ability to determine the state of the system without any influence on its subsequent dynamics, it is impossible to draw any clear conclusion. Recently, the violation of LG inequalities was realized in an optical system with CNOT gate[6] which is a useful method. There are also several other approaches that take advantage of weak measurement[6–8] in which the dynamics of the system are slightly disturbed. In this work, we take a different approach. By replacing the noninvasive measurement assumption with the stationarity assumption,[9,10] we derive testable LG inequalities that have been tested recently in a diamond defect center.[11]

Considering an observable \( Q(t) \) of a two-level system such as the system for the polarization of the photon. We can define \( Q(t) = 1 \) when the state of the photon is \( |H\rangle \), and \( Q(t) = -1 \) when the state of the photon is \( |V\rangle \). The autocorrelation function of this observable is defined as \( K(t_1, t_2) = \langle Q(t_1)Q(t_2) \rangle \). For three different time points \( t_1, t_2 \) and \( t_3 \) \( (t_1 < t_2 < t_3) \), we obtain the two LG inequalities under the realistic description[1]

\[
\begin{align*}
K(t_1, t_2) + K(t_2, t_3) + K(t_1, t_3) & \geq -1, \quad (1) \\
K(t_1, t_3) - K(t_1, t_2) - K(t_2, t_3) & \geq -1. \quad (2)
\end{align*}
\]

Once the assumption of stationarity is introduced, in the case of \( t_2 - t_1 = t_3 - t_2 = t \), the inequalities become[9]

\[
\begin{align*}
K_+ = K(2t) + 2K(t) & \geq -1, \quad (3) \\
K_- = K(2t) - 2K(t) & \geq -1. \quad (4)
\end{align*}
\]

These inequalities set the boundary of the temporal correlations and are amenable to testing in a two-shot experiment.

In this Letter, we compare the two types of LG inequalities and discuss the violation of LG inequalities under the effects of the fine-structure splitting, background noise and temperature in a quantum dot system.

Semiconductor quantum dots, often referred to as “artificial atoms”, have well defined discrete energy levels[12] owing to their three-dimensional confinement of electrons. The atom-like properties of the single semiconductor quantum dot together with the ease of integration into more complicated device structures have made the quantum dot an attractive and widely-studied system for applications in quantum information.[13] One of its potentially useful properties is the ability to emit polarization entangled photon pairs through the radiative decay of the biexciton state.[14–17] This process is presented in Figure 1; a single QD is initially excited to the biexciton state, and subsequently evolves freely. A biexciton photon \( H_{XX} \) or \( V_{XX} \) is emitted as the dot decays to an exciton state by recombining one electron and one hole.

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The polarization of the biexciton photon is either horizontal or vertical, in accordance with the decay into the exciton state $X_H$ or $X_V$; after some time delay $\tau$, the other electron and hole recombine to emit an exciton photon $H_X$ or $V_X$ with the same polarization as that of the earlier biexciton photon. Therefore, this process generates the entangled two-photon state\[^{[18]}\]

$$\left| \Psi \right> = \frac{1}{\sqrt{2}} \left( |H_{XX}H_X + e^{i S \tau} |V_{XX}V_X \right) ,$$

where $S$ is the energy splitting between the exciton states, as shown in Fig. 1, which has conventionally been called the “fine-structure splitting” (FSS).

### Fig. 1. (Color online) Energy-level schematic diagram of the biexciton cascade process. The ground state is $|0\rangle$, the two linear-polarized exciton states are $X_H$ ($|2\rangle$) and $X_V$ ($|1\rangle$), the biexciton state is $XX$ ($|3\rangle$); the fine-structure splitting is $S$ and the spontaneous emission process is denoted by $\gamma_{11}$. A biexciton photon $H_{XX}$ or $V_{XX}$ is emitted as the dot decays to an exciton state by recombining one electron and one hole; after some time delay, the other electron and hole recombine to emit an exciton photon $H_X$ or $V_X$ with the same polarization as that of the earlier biexciton photon.

Taking the acoustic phonon-assisted transition process into account, the density matrix of this four-level system can be described with the master equation\[^{[10]}\]

$$\dot{\rho} = -i[\hat{H}_0, \rho] + L(\hat{\rho}),$$

where

$$\hat{H}_0 = \sum_{i=0}^{3} \omega_i |i\rangle\langle i|.$$  \hspace{1cm} (7)

The $L(\hat{\rho})$ is the Lindblad term denoting dissipation presented in Ref\[^{[20]}\]. In this term, $\gamma_{11}$ and $\gamma_{12}$ denote the photon-assisted transition rates between $|1\rangle$ and $|2\rangle$. The photon absorption rate of the state $|1\rangle$ is $\gamma_{11} = \kappa N_B$ and the emission rate of $|2\rangle$ is $\gamma_{21} = \kappa (N_B + 1)$, where $\kappa$ is the photon-QD interaction rate, which is approximately proportional to the cube of the energy splitting $S$. $N_B$ represents the Bose distribution function of a photon with energy $S$, $N_B = [\exp(S/k_B T) - 1]^{-1}$. In experiment, in order to overcome the influence of FSS, we detect the photon pairs with delays $\tau$ in the range $t \leq \tau \leq (t + \omega)$, by employing a single timing gate $\omega$ \[^{[13]}\] where $t$ is the start time of the gate.

In our model, we assume that the total polarization density matrix $\hat{\rho}_{\text{tot}}$ of the cascaded emission photon includes three parts:\[^{[20]}\] the part with nonclassical correlations $\hat{\rho}_{\text{pol}}$ whose portion $\eta$ is determined by the spectrum overlap of exciton and biexciton photons, the distinguished part $\hat{\rho}_{\text{non}}$, and the background noise $\hat{\rho}_{\text{noise}}$ whose portion is $g$. Therefore,

$$\hat{\rho}_{\text{tot}} = \frac{1}{1 + g} [\eta \hat{\rho}_{\text{pol}} + (1 - \eta) \hat{\rho}_{\text{non}} + g \hat{\rho}_{\text{noise}}].$$

The elements of $\hat{\rho}_{\text{pol}}$ can be derived by the master equation method and quantum regression theorem presented in Ref.\[^{[20]}\]. Therefore the polarization density matrix $\hat{\rho}_{\text{pol}}$ can be calculated from Eqs. (6) and (7) and all nondiagonal terms except $\rho_{14}$ and $\rho_{11}$ are zero. The physical reason for this is that the Hamiltonian in Eq. (6) only couples states that share the same excitation labels $H$ and $V$. The second term $\hat{\rho}_{\text{non}}$ has the same diagonal elements as $\hat{\rho}_{\text{pol}}$, but all its nondiagonal elements are zero. The noise term $\rho_{\text{noise}}$ is set as an identity matrix. Therefore, we can calculate the elements of the total density matrix; in our model, the values of the parameters that we used come from Ref.\[^{[20]}\] and the elements of the total density matrix can be expressed as

$$\hat{\rho}_{\text{tot}} = \begin{pmatrix}
\rho_{11} & 0 & 0 & \rho_{14} \\
0 & \rho_{22} & 0 & 0 \\
0 & 0 & \rho_{33} & 0 \\
\rho_{11} & 0 & 0 & \rho_{44}
\end{pmatrix}. \hspace{1cm} (9)$$

In this study, we consider the two exciton states $|2\rangle$ and $|1\rangle$ in a quantum dot. We define $Q(t) = 1$ when the quantum dot state is $\frac{1}{\sqrt{2}}(|2\rangle + |1\rangle)$ and $Q(t) = -1$ when the quantum dot state is $\frac{1}{\sqrt{2}}(|2\rangle - |1\rangle)$. The quantum dot is initially excited to the biexciton state by a short-pulsed laser and then evolves in a thermal bath. After emitting a biexciton photon, the proposed single quantum dot system will be in an entangled photon-exciton state. For an ideal quantum dot with degenerate intermediate exciton states $|2\rangle$ and $|1\rangle$, it is a maximum entangled state $|\psi\rangle = \frac{\sqrt{2}}{2} (|H\rangle|2\rangle + |V\rangle|1\rangle)$. Then if we find the biexciton photon in state $\frac{\sqrt{2}}{2} (|H\rangle + |V\rangle)$, the state of the quantum dot system will be $\frac{1}{\sqrt{2}} (|2\rangle + |1\rangle)$. With the help of measurement, we can prepare the initial state of the quantum dot system. After a period of time evolution, the quantum dot system will emit the second exciton photon. Similarly, if the state of the second photon is detected to be $\frac{1}{\sqrt{2}} (|H\rangle - |V\rangle)$, it indicates that the state of the quantum dot system is $\frac{1}{\sqrt{2}} (|2\rangle - |1\rangle)$ at that moment. In this case, because of the relationship between the photon and the quantum dot system, we can detect the photon state to evaluate the LG inequalities instead of detecting the state of the quantum dot system. Therefore, we can evaluate the LG inequalities with these photon states. We then define the observable $Q(t) = 1$ when the state of the photon is $|+\rangle = \frac{\sqrt{2}}{2} (|H\rangle + |V\rangle)$, and $Q(t) = -1$ when the state
of the photon is $|−⟩ = \frac{1}{\sqrt{2}}(|H⟩ − |V⟩)$. At the time $t = 0$, we detect the first photon under the bases $|+⟩$ and postselect the result of $|+⟩$ to prepare the initial state of the quantum dot system. After a period of time $t$, we detect the second photon under the bases $|+⟩$ and $|−⟩$. We have the joint probability $P_{++}$ when the state of the second photon is $|+⟩$ and the probability $P_{+-}$ when the state of the second photon is $|−⟩$. After normalization, the autocorrelation can be expressed as

$$P_{++}(t) = ⟨Ψ_{1}|ρ_{rot}|Ψ_{1}⟩, P_{+-}(t) = ⟨Ψ_{2}|ρ_{rot}|Ψ_{2}⟩,$$  

$$K(t) = \frac{P_{++}(t) − P_{+-}(t)}{P_{++}(t) + P_{+-}(t)},$$

where $|Ψ_{1}⟩ = |+⟩ \otimes |+⟩, |Ψ_{2}⟩ = |+⟩ \otimes |−⟩$. We then obtain the final autocorrelation

$$K(t) = \frac{ρ_{14}(t) + ρ_{41}(t)}{ρ_{11}(t) + ρ_{22}(t) + ρ_{33}(t) + ρ_{44}(t)}.$$  

Therefore, we can evaluate the LG inequalities through the expressions $K_+ = K(2t) + 2K(t)$ and $K_- = K(2t) − 2K(t)$. The autocorrelation will be measured twice by two independent experiments that begin with a primary system in an identical initial state which evolves under identical conditions. In the first experiment, the measurement of the autocorrelation is made at the time $t$ and the second measurement is made at the time $2t$ by another experiment and the joint probability can be calculated through the total density matrix of the photon pairs.

![Fig. 2.](image_url) Fig. 2. Two types of LG inequalities. The fine-structure splitting $S = 3\mu$eV, background noise $g = 0$, the gate width $ω = 50$ ps and the temperature $T = 5$ K.

Our result is shown in Fig. 2. We can see that the curve of the LG oscillates with time, what’s more, the amplitude of the curve decays as the time passes. By comparing the two types of LG inequalities, we find that the $K_-$ reaches the classical limit $−1$ first. As we know, when the time delay of the photon pairs increases, the biphoton coincidence decreases. Therefore, $K_-$ is easier to be tested in the experiment and we discuss the following results for $K_-.$

We analyse the relationship between the LG inequality and the fine-structure splitting. The result is presented in Fig. 3. In this case, the background noise $g = 0.3$, the gate width $ω = 50$ ps and the temperature $T = 5$ K. When the fine-structure splitting is small, the LG inequality is violated easily. However, with an increase in the fine-structure splitting, the violation of the LG inequality becomes increasingly weak. It is seen that $K_-$ does not violate the classical limit $−1$ when the fine-structure splitting is sufficiently large. This implies that when the fine-structure splitting becomes large, the evolution process can be described by classical realistic theory. In order to violate the Leggett–Garg inequalities, there must be some correlations between the states of a single system at different times. From Eq. (5), we can see clearly that the phase of the superposition rotates as the intensity decays in a quantum dot with finite splitting; after some time delay, instantaneous superpositions largely cancel out with those at other times with opposing phase, giving rise to more classical photon pairs.

![Fig. 3.](image_url) Fig. 3. Relationship between fine-structure splitting and the Leggett–Garg inequality. Under the conditions of the background noise $g = 0.3$, the gate width $ω = 50$ ps and the temperature $T = 5$ K. The violation of the LG inequality becomes easier as the fine-structure splitting decreases.

![Fig. 4.](image_url) Fig. 4. Relationship between background noise and the LG inequality. The fine-structure splitting $S = 3\mu$eV, the gate width $ω = 50$ ps and the temperature $T = 5$ K. Furthermore, the influence of background noise and temperature are investigated. As shown in Figs. 4 and 5, when the background noise and temperature decrease, the curve of the LG inequality falls below the classical limit $−1$ easily. The transition from a quantum process to a classical process has also been seen clearly. Nowadays, the difficulty of superposition of macroscopic states is explained by decoherence, where the superposition of distinct states is destroyed by coupling with unwanted degrees of freedom. When the influence of the environment increases, it becomes
difficult to violate the Leggett–Garg inequalities.

Finally it should be noticed that there is an approximation in our model. We use the state of the photon emitted to determine the state of the quantum dot and this approach is valid when the fine structure splitting is zero. In a real situation, we need the fine structure splitting to be nonzero in order to introduce the evolution and the partial distinguishability of spectra between $|H\rangle$ and $|V\rangle$ will introduce errors. However, it can be easily verified that the errors only make the LG inequality more difficult to be violated, i.e., the requirement is not loosened in our model.

In summary, we have investigated the violation of the LG inequalities in a quantum dot system. When the fine-structure splitting of the quantum dot system, background noise and the temperature become smaller, we achieve the maximal violation. Better results may be obtained when we improve the method by which we obtain the initial state. Finally, we can see clearly from the results that the LG inequalities can be used as a criterion to identify the boundary of the classical realistic description.

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