

Dephasing of electrons by two-level defects in quantum dots

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The electron dephasing time τ_ϕ in a diffusive quantum dot is calculated by considering the interaction between the electron and dynamical defects, modeled as two level systems. Using the standard tunneling model of glasses, we obtain a linear temperature dependence of $1/\tau_\phi$, consistent with the experimental observation. However, we find that, in order to obtain dephasing times on the order of nanoseconds, the number of two-level defects needs to be substantially larger than the typical concentration in glasses. We also find a finite system-size dependence of τ_ϕ , which can be used to probe the effectiveness of surface-aggregated defects.

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I. INTRODUCTION

Interference of the electron's paths in a mesoscopic conductor results in various quantum phenomena such as the universal conductance fluctuation, persistent current, and weak localization. In all of these phenomena, the dephasing time τ_ϕ appears as a typical time scale over which the electronic trajectories have interference; weak-localization correction to conductivity—for example, is conventionally used for the experimental determination of the dephasing time.¹ In the moderate temperature range, experimentally determined² values of τ_ϕ in diffusive metals are found to be in excellent agreement with the theoretical predictions of τ_ϕ due to electron-electron interaction.¹ While it is theoretically expected that $\tau_\phi \rightarrow \infty$ as $T \rightarrow 0$ in the absence of other external sources of dephasing, τ_ϕ is found to saturate at low temperatures in *almost* all experiments,³ including the recent carefully performed experiments.⁴⁻⁷ This severe discrepancy between theory and experimental observation of low-temperature saturation has fast become a topic of controversy^{8,9} surrounding the question whether the idea⁴ and the theory⁸ of zero-point fluctuations of the electromagnetic field created by the electron-electron interaction as a source of dephasing are tenable on general grounds. This poses a serious problem as zero-temperature dephasing of electrons has been argued to be relevant to the problems of persistent current in normal metals,¹⁰ the low-temperature metal/insulator, quantum-Hall/insulator and superconductor/insulator transitions,¹¹⁻¹³ and transport through various normal-metal/superconductor hybrid junctions;^{14,15} but the most unsettling consequence is the negation of the fundamental premise upon which the theories—and hence our understanding—of metals and insulators are based: the many-body Fermi-liquid picture.

Among various sustained efforts to find a zero-temperature dephasing mechanism other than electron-electron interaction, dephasing due to dynamical defects inside the conductor has been recently argued^{16,17} to be important to the saturation problem. Low-energy excitations of the dynamical defects are usually modeled by two-level systems (TLS). Invoked some three decades ago, first by

Anderson, Varma and Halperin,¹⁸ and also by Phillips,¹⁹ the tunneling model of TLS has been quite successful in explaining various anomalies in the acoustic, dielectric, and thermodynamic properties of structural glasses and other amorphous solids.²⁰

Imry, Fukuyama, and Schwab¹⁶ have recently suggested that the saturation behavior may have the same origin as the $1/f$ conductance noise, arising from the two-level defects. Zawadowski, von Delft, and Ralph¹⁷ have argued that the apparent saturation of τ_ϕ may be caused by the two-channel Kondo effect due to electron-TLS scatterings. However, it was pointed out^{11,5} that hysteresis or switching behavior, expected from the effects of TLS, was not observed in experiments. In addition, various concentration-dependent Kondo-like bulk trends anticipated in these theories were also not observed in the experiments.

In this paper, we investigate the role of two-level defects in the dephasing of electrons in *quantum dots*. In the recent experiments from the Marcus group, Huibers and coworkers have observed the saturation of dephasing time in open quantum dots²¹ below 0.1 K along with a strong temperature dependence above 0.1 K. In addition to this experiment, saturation of τ_ϕ in quantum dots has also been reported in other experiments.^{22,23} If one assumes that two-level defects are responsible for the saturation of dephasing time in these experiments,²¹ it is then natural to suppose just above 0.1 K the linear temperature dependence should be explained by two-level defects as well. Our calculations indeed show that the dephasing rate due to the TLS does have a linear temperature dependence. However, we find that the magnitude of the dephasing by two-level defects is too small to explain the experimentally observed dephasing time of nanoseconds. This implies that either other mechanisms are more effective or surface-aggregated two-level defects play a dominant role; defects on a disordered surface are likely to have unusual distributions in their splitting energies. We suggest that the surface defects can be experimentally probed by measuring the size dependence of dephasing time.

Consideration of two-level defects in quantum dots for their dominant role in dephasing is motivated by the experimentally observed tell-tale signs of TLS in quantum dots:

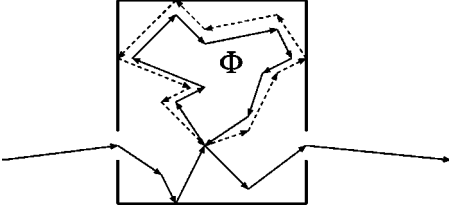


FIG. 1. A schematic figure of the electron interferometry involving a quantum dot that is studied in this paper.

hysteresis and switching behavior, which have been in fact detected in various quantum-dot experiments^{24–26} unlike in the experiments on higher-dimensional diffusive metals. In a quantum dot, usually the Thouless energy E_T is the largest energy scale unlike the diffusive metallic case.^{1,8} Therefore, the results obtained in the diffusive metallic cases^{1,8} cannot be applied (even after the appropriate dimensional considerations) to quantum dots. Thouless energy is defined by $E_T \equiv \hbar D/L^2$, where D is the diffusion constant and L is the typical system size.

Dephasing generally describes the loss of coherence or suppression of interference. Hence, it is important to know which kind of paths are considered before defining the typical time scale of the loss of interference along these paths. In this paper, we are concerned with pairs of time-reversed paths that return to the origin in a diffusive system, shown in Fig. 1. These time-reversed paths enclose magnetic flux; their interference manifests in the weak-localization correction to conductivity. These paths are chosen for the problem at hand, because their contribution to conductivity does not vanish even after disorder averaging. In the interferometry studied in this paper, change of the mean conductance at a finite field from its zero field value, $\delta g = \langle \langle g \rangle \rangle_{B \neq 0} - \langle \langle g \rangle \rangle_{B=0}$, is used to extract the dephasing time from experimental data, where $\langle \langle \dots \rangle \rangle$ means disorder averaging. Using the phenomenological random matrix theory (see, for example, Refs. 27,28), τ_ϕ can be defined—for instance, by the formula

$$\delta g \approx \frac{e^2}{h} \left(\frac{N}{2N + \frac{2\pi\hbar}{\tau_\phi \Delta}} \right), \quad (1)$$

where N is the number of channels connected to the quantum dot. Although, the formulas for the conductance change are model dependent,^{27,28} the difference in the equations in these models is not significant for the interpretation of τ_ϕ measured in experiments.²¹ In this paper, we will refer to τ_ϕ obtained from the measured δg as in Ref. 21, without discussing how δg is related to τ_ϕ any further.

Our calculation of dephasing time is similar to the general approach of Stern, Aharonov, and Imry.²⁹ Based on the interference of two time-reversed trajectories, we calculate a typical time scale over which the environmental state remains in the initial state. Dephasing rate $1/\tau_\phi$ due to electron-electron interactions in diffusive quantum dots has been calculated by Sivan and coworkers.³⁰

$$\frac{1}{\tau_\phi} \Big|_{e-e} \sim \delta_1 \left(\frac{\epsilon}{E_T} \right)^2, \quad \epsilon \gg k_B T; \quad (2)$$

where ϵ is the excitation energy of the particle and δ_1 is the mean level spacing. However, it should be noted that the direct application of Eq. (2) to experimental data²¹ is difficult, because it is not meaningful to estimate the temperature dependence of τ_ϕ by merely replacing ϵ with $\sim k_B T$ in Eq. (2).³¹

The organization of the paper is as follows. In Sec. II, we describe how dephasing time is calculated in a general framework. In Sec. III, interaction between the two-level defects and electrons is discussed. In Sec. IV, we show the calculation of $1/\tau_\phi$ in the presence of two-level defects with widely and narrowly distributed energies. We conclude in Sec. V.

II. INTERFERENCE AND DEPHASING OF PARTICLE'S TRAJECTORY

Let us consider the event that the electron is at the position \mathbf{r} at an initial time $t=0$, and it arrives at the position \mathbf{r}' by diffusive motion after a time τ_0 . The environmental state changes from η to η' in this process; the corresponding probability amplitude of the event is $\rho(\mathbf{r}', \mathbf{r}, \eta', \eta; \tau_0)$.

The description of the suppression of interference in electron's paths by the electron-TLS interaction can be considered in two different approaches:

- (i) The electron in the two different paths produces two different time-dependent electric fields on TLS, thereby, TLS go to different states, which suppresses interference.
- (ii) The fluctuating dipole moment of TLS produces the time-dependent electric field, thereby, the electron in the two different paths gains random phase, which also suppresses interference.

In general, these two approaches are not equivalent, because the presence of the electron induces a back reaction from the TLS environment. However, in the presence of weak interaction between the particle and the environment, it is known that either the two descriptions are equivalent, or at least they give the same dephasing rate up to the second order in the interaction.³² In this paper, we use approach (i). Following the scheme of Chakravarty and Schmid,³³ we use semiclassical approximation on particle's trajectory and we consider quantum-mechanical evolution of the TLS (environment) states. We further assume that the TLS environment does not influence the classical paths of the electron, therefore, the diffusive electron motion comes from only static disorder. Under certain conditions, the two-level defects might be able to effectively change the semiclassical paths of the electrons. In that case, one may estimate τ_ϕ by calculating electron-TLS inelastic-scattering time. However, τ_ϕ begins to lose its meaning as a dephasing time, since we lose the semiclassical picture of the electron's path.

We describe the tunneling motion in the TLS environment in a fully quantum-mechanical way. To this end, we consider the time-dependent potential $\hat{V}[\mathbf{r}(t)]$ exerted by the moving electron of the path $\mathbf{r}(t)$ on a two-level defect. The probability amplitude is given by

$$\rho(\mathbf{r}', \mathbf{r}, \boldsymbol{\eta}', \boldsymbol{\eta}; \tau_0) = \sum_j A_j(\mathbf{r}', \mathbf{r}; \tau_0) e^{iS_j} \langle \boldsymbol{\eta}' | U_j(\tau_0) | \boldsymbol{\eta} \rangle, \quad (3)$$

where A_j and S_j are the corresponding amplitude and action of a classical electron's trajectory labeled by j . $U_j(\tau_0)$ is a time-evolution operator (in the interaction picture) of the environmental state associated with the electron trajectory $\mathbf{r}_j(t)$

$$U_j(\tau_0) = \hat{T} \exp \left[\frac{i}{\hbar} \int_0^{\tau_0} V_I[\mathbf{r}_j(t), t] dt \right], \quad (4)$$

where \hat{T} is the time-ordering operator and

$$\hat{V}_I[\mathbf{r}_j(t), t] = e^{(i/\hbar)H_{env}t} \hat{V}[\mathbf{r}_j(t)] e^{-(i/\hbar)H_{env}t}. \quad (5)$$

The probability $P(\mathbf{r}', \mathbf{r}, \boldsymbol{\eta}; \tau_0)$ of finding the particle at \mathbf{r}' after time τ_0 , initially at \mathbf{r} with the environment in the initial state $|\boldsymbol{\eta}\rangle$, is given by the sum of the absolute square of the probability amplitudes over the final states of the environment;

$$\begin{aligned} P_{\boldsymbol{\eta}}(\mathbf{r}', \mathbf{r}; \tau_0) &= \int d\boldsymbol{\eta}' |\rho(\mathbf{r}', \mathbf{r}, \boldsymbol{\eta}', \boldsymbol{\eta}; \tau_0)|^2 \quad (6) \\ &= \sum_j |A_j(\mathbf{r}', \mathbf{r}; \tau_0)|^2 \\ &\quad + \sum_{j \neq k} A_j A_k^* e^{i(S_j - S_k)} \\ &\quad \times \langle \boldsymbol{\eta} | U_k^\dagger(\tau_0) U_j(\tau_0) | \boldsymbol{\eta} \rangle, \quad (7) \end{aligned}$$

using the completeness relation for the environmental states.

The return probability $P_{\mathbf{r}, \boldsymbol{\eta}}(\tau_0)$ of the electron is defined by the probability of finding the electron at position \mathbf{r} after time τ_0 , initially at the same position with the environmental state $|\boldsymbol{\eta}\rangle$;

$$P_{\mathbf{r}, \boldsymbol{\eta}}(\tau_0) = P_{\boldsymbol{\eta}}(\mathbf{r}', \mathbf{r}; \tau_0) |_{\mathbf{r}' \rightarrow \mathbf{r}} \quad (8)$$

$$\begin{aligned} &= \sum_j |A_j(\mathbf{r}, \mathbf{r}; \tau_0)|^2 + \sum_j |A_j|^2 \\ &\quad \times \left\langle \boldsymbol{\eta} \left| \frac{U_{j^T}^\dagger U_j + U_j^\dagger U_{j^T}}{2} \right| \boldsymbol{\eta} \right\rangle + \dots, \quad (9) \end{aligned}$$

where j^T denotes the time-reversed path of j . The first term in Eq. (9) is termed as the classical return probability $P_{\mathbf{r}}^{class}(\tau_0)$. The second term comes from the interference of the pair of time-reversed paths. The remaining terms, which do not appear in the above equation, vanish upon ensemble averaging over disorder due to the random differences in its classical action $S_j - S_k$ for $k \neq j, j^T$. The coherent part $P_{\mathbf{r}, \boldsymbol{\eta}}^{coh}(\tau_0)$ of the return probability is the second term in Eq. (9);

$$P_{\mathbf{r}, \boldsymbol{\eta}}^{coh}(\tau_0) = \sum_j |A_j(\mathbf{r}, \mathbf{r}; \tau_0)|^2 \text{Re} \langle \boldsymbol{\eta} | U_j^\dagger(\tau_0) U_{j^T}(\tau_0) | \boldsymbol{\eta} \rangle. \quad (10)$$

Now, the dephasing time can be defined as a time scale for $P^{coh}(\tau_0)$ to vanish with a decreasing function such as $\exp(-\tau_0/\tau_\phi)$. But for the present purpose, the particular exponential form of time dependence is not needed.

III. ELECTRON-TLS INTERACTION

We consider two-level tunneling systems (TLS) (Ref. 20) as the environment for an electron in the quantum dot. Let us first consider TLS's that have asymmetry energy Δ and the tunnel splitting energy Δ_0 . The TLS's are assumed to be randomly distributed over the dot with their electric dipole moments randomly oriented. We will assume the dipole moment is not too strong so that we do not have to consider interaction among the TLS. The density of the TLS will be assumed to be not too high so that multiple-scattering events between the electron and the dipoles can be neglected. Within these approximations, we calculate the return probability of an electron in the presence of a single TLS, thereby, we extend the results to the case of many randomly distributed TLS's. The Hamiltonian of the TLS can be written in terms of the localized wave functions of the double well potential and also in terms of the eigenenergy basis

$$H_{TLS} = \frac{1}{2} \begin{pmatrix} \Delta & \Delta_0 \\ \Delta_0 & -\Delta \end{pmatrix} \rightarrow \frac{1}{2} \begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix}, \quad (11)$$

where $E = \sqrt{\Delta^2 + \Delta_0^2}$ and the transformation denoted by the arrow means localized wave-function representation \rightarrow eigen wave-function representation. The dipole strength operator \hat{p} is defined in the eigen wave-function representation:

$$\hat{p} = p_0 \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \rightarrow p_0 \begin{pmatrix} \Delta/E & \Delta_0/E \\ \Delta_0/E & -\Delta/E \end{pmatrix}, \quad (12)$$

where p_0 is the dipole moment when the particle is located in one of the wells of the defect potential. In the following section, we will use the eigen wave-function representation in which H_{TLS} is diagonal. The TLS Hamiltonian will be used for the environment Hamiltonian $H_{env} = H_{TLS}$.

TLS dipole at the position \mathbf{R} feels the electric field $\mathbf{E}(\mathbf{R})$ produced by the moving electron. The resulting interaction energy can be expressed by the operator $\hat{V}[\mathbf{r}_j(t)]$

$$\hat{V}[\mathbf{r}_j(t)] = -\hat{p} \cdot \mathbf{E}(\mathbf{R}) = -\hat{p} \cdot \nabla_{\mathbf{R}} V_c[\mathbf{R} - \mathbf{r}_j(t)], \quad (13)$$

where \hat{p} is the dipole moment operator for the TLS, which is along the direction of unit vector \mathbf{n} . V_c is Coulomb interaction potential

$$V_c[\mathbf{R} - \mathbf{r}_j(t)] = \frac{e}{\epsilon^* |\mathbf{R} - \mathbf{r}_j(t)|} \approx \frac{1}{L^d} \sum_{\mathbf{q}} v_{\mathbf{q}} e^{i\mathbf{q} \cdot (\mathbf{r}_j - \mathbf{R})}, \quad (14)$$

where e is the electric charge, ϵ^* is the dielectric constant of the dot material, L is the linear system size, and d is the spatial dimensionality of the dot ($d=2,3$). Here, $v_{\mathbf{q}}$ is the finite Fourier transform of Coulomb potential

$$v_{\mathbf{q}} = \int_{dot} d\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} \left(\frac{e}{\epsilon^* |\mathbf{r}|} \right) \quad (15)$$

$$= \frac{2\pi e}{\epsilon^* q} \quad (2D, \quad q \neq 0) \quad (16)$$

$$= \frac{4\pi e}{\epsilon^* q^2} \quad (3D, \quad q \neq 0). \quad (17)$$

We use the discrete values of $\mathbf{q} = 2\pi/L(m, n, k)$ for the three-dimensional case, and $\mathbf{q} = 2\pi/L(m, n)$ for the two-dimensional case, where l, m , and n are integers. By inserting Eq. (14) into Eq. (13), we get

$$\hat{V}[\mathbf{r}_j(t)] = \frac{1}{L^d} \hat{p} \sum_{\mathbf{q}} (i\mathbf{n} \cdot \mathbf{q} v_{\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}}) e^{i\mathbf{q}\cdot\mathbf{r}_j(t)}. \quad (18)$$

Derivation of the Eq. (18) is based on the semiclassical approximation of the electron's motion and the unscreened dipole moment of TLS. From a purely quantum-mechanical point of view, one can consider the TLS-electron interaction similar to its treatment in metallic glasses.³⁵ When the TLS has two positions $\mathbf{R}^{\pm} = \mathbf{R} \pm \mathbf{d}/2$, the pure quantum-mechanical TLS-electron interaction \hat{V}_{QM} is written as³⁵

$$\hat{V}_{QM} = \frac{1}{L^d} \frac{\hat{p}}{p_0} \sum_{\mathbf{q}} i\mu_{\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}} \sin(\mathbf{q}\cdot\mathbf{d}/2) \sum_k c_k^{\dagger} c_{k+\mathbf{q}}, \quad (19)$$

where c_k^{\dagger} (c_k) is the electron creation(annihilation) operator with momentum k , and $\mu_{\mathbf{q}}$ is the Fourier transform of the ionic potential. However, the pure quantum-mechanical approach is not reliable, because the concept of dephasing becomes ambiguous as we leave the semiclassical approximation, which has been pointed out in Ref. 34. Here, we merely get some useful information by comparing the quantum-mechanical Hamiltonian in Eq. (19) to our semiclassical potential in Eq. (18). Since $\mathbf{q}\cdot\mathbf{d} \ll 1$ in quantum dots (this is true—generally speaking, when the Fermi wavelength is much larger than d), $\sin(\mathbf{q}\cdot\mathbf{d}/2) \approx \mathbf{q}\cdot\mathbf{d}/2 = \mathbf{n}\cdot\mathbf{q}p_0/e$. Equation (19) can be understood as the interaction between the TLS-dipole coupled to an effective electric field produced by the electron.

One of the two differences between Eqs. (18) and (19) is that the electron interacts with the ion through a screened interaction $\mu_{\mathbf{q}}$ in Eq. (19) rather than the direct Coulomb interaction $v_{\mathbf{q}}$, as in Eq. (18). The specific form of $\mu_{\mathbf{q}}$ is not known though it is expected to be less than $v_{\mathbf{q}}$. If a screened interaction $\mu_{\mathbf{q}}$ is used, then the calculated dephasing rate $1/\tau_{\phi}$ would be smaller than that with the unscreened interaction $v_{\mathbf{q}}$. In this paper, we will use $v_{\mathbf{q}}$ instead of $\mu_{\mathbf{q}}$. The second difference, which is rather important, is the quantum-

mechanical nature of the electron motion in Eq. (19). $\hat{V}[\mathbf{r}_j(t)]$ in Eq. (18) is understood as $\langle \psi_j(\mathbf{r}, t) | \hat{V}_{QM} | \psi_j(\mathbf{r}, t) \rangle$, where $\psi_j(\mathbf{r}, t)$ is the time-dependent wave function that describes a wave packet corresponding to the trajectory j ,

$$e^{i\mathbf{q}\cdot\mathbf{r}_j(t)} \approx \left\langle \psi_j(\mathbf{r}, t) \left| \sum_k c_k^{\dagger} c_{k+\mathbf{q}} \right| \psi_j(\mathbf{r}, t) \right\rangle. \quad (20)$$

At a finite temperature T , the wave-packet state $|\psi_j(\mathbf{r}, t)\rangle$ will consist of mostly the eigenenergy states with energies limited within $\epsilon_F \pm k_B T$. Therefore, the time dependence of $e^{i\mathbf{q}\cdot\mathbf{r}_j(t)}$ will be limited by the frequency window $|\omega| < k_B T$. The frequency cutoff will be performed in our semiclassical calculation later.

IV. DEPHASING RATE

Let us consider a quantum dot as a rectangular box with volume $\mathcal{V} = L \times L \times L$ (L or $a \ll L$ for 2D dots), and a diffusion constant D . Using Eq.s (4), (10), and (18), we get the coherent part of the return probability $P^{coh}(\tau_0)$ of the electron, initially at $\mathbf{r}=0$ (details of the derivation is given in the appendix);

$$P^{coh}(\tau_0) = \frac{1}{\mathcal{V}} \left[1 + \sum_{\mathbf{q}} \frac{|\langle + | \hat{p} | - \rangle|^2 q^2 v_{\mathbf{q}}^2}{3\hbar^2 L^{2d}} \int_0^{\tau_0} dt^+ \int_{-t^+}^{t^+} dt^- \right. \\ \left. \times \{ \cos \Omega(t^+ - \tau_0) - \cos \Omega t^- \} \frac{1}{2\pi} \right. \\ \left. \times \int d\omega \frac{\exp(i\omega|t^-|)}{i\omega + Dq^2} \right], \quad (21)$$

where $\Omega = 1/\hbar \sqrt{\Delta^2 + \Delta_0^2}$. To simplify the calculation, from here onward, we consider the return probability of the particle at the origin $\mathbf{r}=0$.

The frequency of the time-dependent electric field produced by the electron is not infinitely large; it has an upper cutoff. By assuming the electron to be in equilibrium with other electrons at temperature T , the high-frequency cutoff of ω is given by $k_B T$ ($|\omega| < k_B T$); this is true only at temperatures that are not too low. Note that because of the finite size of the system, $qv_{\mathbf{q}} = 0$ for $q=0$. Therefore, there is no divergence at low frequencies and the low-frequency cutoff of ω does not play an important role. By integrating Eq. (21) with the condition $|\omega| < k_B T$, we obtain the coherent part of the return probability P^{coh} , which decays as $\propto 1 - \tau_0/\tau_{\phi} + \dots$. We have now defined τ_{ϕ} as the dephasing time. The dephasing rate $1/\tau_{\phi}$ from a randomly distributed TLS with an asymmetry energy Δ and a tunnel splitting Δ_0 is given by

$$\frac{1}{\tau_{\phi}(\Delta, \Delta_0)} = \frac{2|\langle + | \hat{p} | - \rangle|^2}{3\hbar^2 L^{2d}} \sum_{\mathbf{q}} q^2 v_{\mathbf{q}}^2 \frac{Dq^2}{\Omega^2 + (Dq^2)^2} \quad (22)$$

$$\approx \frac{2p_0^2 \Delta_0^2}{3\hbar^2 D(\Delta^2 + \Delta_0^2)} \sum_{0 < |\mathbf{q}| < \pi/l} v_{\mathbf{q}}^2 L^{-2d}. \quad (23)$$

We have used $\Omega\tau_D \ll 1$ in the second equality in Eq. (23). $1/\tau_\phi$ obtained above is valid only when $k_B T > \hbar\Omega$, while in the other case, $P^{coh}(\tau_0)$ is an oscillating function of τ_0 with a small amplitude. Therefore, $1/\tau_\phi$ ($k_B T < \hbar\Omega$) is negligible.

By inserting Eqs. (16) and (17) into Eq. (23), when $k_B T > \sqrt{\Delta^2 + \Delta_0^2}$, we get

$$\frac{1}{\tau_\phi(\Delta, \Delta_0)} = \frac{2p_0^2 e^2 \Xi_d \Delta_0^2}{3\hbar^2 D \epsilon^{*2} L^2 (\Delta^2 + \Delta_0^2)}, \quad (24)$$

where $d=2,3$ is the spatial dimension of the quantum dot, and

$$\Xi_2 = \sum_{0 < m^2 + n^2 < (L/l)^2} \frac{1}{m^2 + n^2}, \quad (25)$$

$$\Xi_3 = \frac{1}{\pi^2} \sum_{0 < m^2 + n^2 + k^2 < (L/l)^2} \frac{1}{m^2 + n^2 + k^2}. \quad (26)$$

A. TLS with widely distributed asymmetry and tunnel-splitting energies

We can generalize $1/\tau_\phi$ to the case where the TLS's are distributed with a distribution function $f(\Delta, \Delta_0)$,

$$\frac{1}{\tau_\phi} = \mathcal{V} \int d\Delta \int d\Delta_0 \frac{1}{\tau_\phi(\Delta, \Delta_0)} f(\Delta, \Delta_0). \quad (27)$$

By inserting Eq. (24) into the above equation, we get

$$\frac{1}{\tau_\phi} = \frac{L^{d-2}}{D} \frac{2p_0^2 e^2 \Xi_d a^{3-d}}{3\hbar^2 \epsilon^{*2}} S(T), \quad (28)$$

where

$$S(T) = \int d\Delta \int d\Delta_0 \frac{\Delta_0^2}{\Delta^2 + \Delta_0^2} f(\Delta, \Delta_0) \theta(k_B T - \sqrt{\Delta^2 + \Delta_0^2}), \quad (29)$$

where a is the thickness of the dot in case $d=2$. It is interesting to note that at $d=2$, the dephasing time does not depend on the dot area.

To calculate τ_ϕ , we use the standard tunneling model for the two-level defects.^{18,19} The essential postulate in this theory is the uniform distribution of the tunneling parameter λ associated with the tunnel splitting $\Delta_0 \propto e^{-\lambda}$. The energy distribution function $f(\Delta, \Delta_0)$ in this case is written as

$$f(\Delta, \Delta_0) = \frac{\bar{P}}{\Delta_0}. \quad (30)$$

Furthermore, it is also assumed that Δ_0 has a nonzero minimum value $\Delta_{0,\min}$. By applying this distribution, we find,

$$S(T) = \bar{P} \int_{\Delta_{0,\min}}^{k_B T} d\Delta_0 \int_{-\sqrt{(k_B T)^2 - \Delta_0^2}}^{\sqrt{(k_B T)^2 - \Delta_0^2}} d\Delta \frac{\Delta_0}{\Delta^2 + \Delta_0^2} \quad (31)$$

$$= 2\bar{P} \Delta_{0,\min} \mathcal{F}(k_B T / \Delta_{0,\min}), \quad (32)$$

where

$$\mathcal{F}(z) = \int_1^z dx \tan^{-1} \frac{\sqrt{z^2 - x^2}}{x}. \quad (33)$$

Here, the above expression is valid for $k_B T < \Delta_{0,\max}$, which is an realistic and common assumption for the temperature below 1 K. Note that $\mathcal{F}(z) \sim z \ln z$ when $z \gg 1$, therefore in the case of $k_B T \gg \Delta_{0,\min}$, we expect the following temperature dependence:

$$\frac{1}{\tau_\phi} \propto T \ln \left(\frac{k_B T}{\Delta_{0,\min}} \right), \quad (34)$$

which is closer to T rather than T^2 .

Now let us estimate τ_ϕ quantitatively. We consider the experiments by Huibers and coworkers²¹ on two-dimensional ballistic semiconductor quantum dots. The quantum dots in the experiments²¹ are in the ballistic regime, while our calculation of τ_ϕ is for diffusive quantum dots. However, since the dephasing time is in the ergodic regime ($\tau_\phi > \tau_D$), the results for diffusive dots should be applicable to the chaotic quantum dots in the ballistic regime. The diffusion coefficient is obtained through the ergodic time scale and $D \sim (E_{Th}/\delta_1)(\hbar/2m^*)$, where m^* is the effective mass of the electron (for GaAs, $m^* = 0.067m_e$), and $E_{Th}/\delta_1 \sim 30$ for Ref. 21. For ballistic dots, the Thouless energy is given by $\hbar v_F/L$. For GaAs, $\hbar^2 \epsilon^*/m^* e^2 \sim 10$ nm. A reasonable size of the dipole moment is $p_0 \sim e \times 10^{-10} m$. The thickness of the two-dimensional quantum dot is roughly $a \sim 10$ nm. By putting together Ξ_2 and $\ln(k_B T / \Delta_{0,\min})$ into Eq. (28), which are roughly on the order of 1–10, we find

$$1/\tau_\phi \sim (10^{-16} - 10^{-15}) m^3 s^{-1} \bar{P} k_B T. \quad (35)$$

In order to obtain $\tau_\phi \sim 1$ ns near $T=0.1$ K, the average concentration should be $\bar{P} \sim (10^{48} - 10^{49}) J^{-1} m^{-3}$. Although this number is not completely unreasonable, it is too large to be expected from well-textured semiconductors used in the experiments.²¹ For comparison, we note that glassy materials possess a typical TLS concentration of $\bar{P} \sim 10^{45} - 10^{46} J^{-1} m^{-3}$.

One may anticipate a different temperature dependence that might show the saturation of τ_ϕ by considering the dissipative two-level system due to TLS-phonon interactions or incoherent two-level systems due to TLS-TLS interactions. However, it is very difficult to expect that the dephasing rate is enhanced by several orders of magnitude by such interactions.

The large magnitude of \bar{P} may be possible if a large enough number of two-level defects aggregate on the surface of the quantum dots. This possibility can be experimentally checked by varying the system size and the dimensionality. Using our results,

$$1/\tau_\phi \propto L^{d-2} a^{3-d}. \quad (36)$$

For example, for a 2D quantum dot, the dephasing rate $1/\tau_\phi$ by ‘‘intrinsic’’ two-level defects will increase as the thickness a of the dot increases, whereas it will decrease with a for surface defects.

B. TLS with a narrow-energy distribution of asymmetry and splitting energies

Low-energy excitations exist in semiconductor crystals due to the tunneling of impurity ions between equivalent interstitial lattice sites. Due to the crystal fields, definite positions are preferred and a wide distribution of excitation energies is not expected; in glasses, the wide distribution arises because of structural disorder. However, defects on the surface may result in a wider distribution of energies because of surface roughness. A single tunnel-splitting energy implies a narrow distribution of relaxation times such that the standard tunneling model, applicable to structural glasses, as discussed in the previous section, is not valid.³⁶

In this section, we consider a well-defined tunnel-splitting energy Δ_0 rather than a wide distribution. The asymmetry may be uniformly distributed with a gaussian width Δ_1 , usually determined from the experimental data. The distribution function is defined as

$$f(\Delta) = n_{TLS} \frac{1}{\Delta_1 \sqrt{\pi}} e^{-\Delta^2/\Delta_1^2}. \quad (37)$$

n_{TLS} is the TLS density.

The function $S(T)$ defined in Eq. (29) in the expression for the dephasing rate $1/\tau_\phi$ is simplified to

$$S(T) = \int d\Delta \frac{\Delta_0^2}{\Delta^2 + \Delta_0^2} f(\Delta) \theta(k_B T - \sqrt{\Delta^2 + \Delta_0^2}). \quad (38)$$

Note that the variable Δ_0 is not integrated over, in contrast to the case for the standard tunneling model; and the final result depends on Δ_0 . Evaluation of the above integral yields

$$S(T) \sim n_{TLS} \quad (k_B T \gg \Delta_0 \gg \Delta_1) \quad (39)$$

$$\sim \frac{\Delta_0}{\Delta_1} n_{TLS} \quad (k_B T \gg \Delta_1 \gg \Delta_0). \quad (40)$$

If temperature is larger than the energy scales of TLS, then it is possible to obtain saturation or temperature-independent dephasing rate $1/\tau_\phi$. In realistic systems, Δ_0 is usually a small fraction of Δ_1 ; thus, the experimentally relevant limit is the second case, $\Delta_1 \gg \Delta_0$, in expression (40). The dephasing rate can now be obtained:

$$\frac{1}{\tau_\phi} \sim (10^{-16} - 10^{-15}) n_{TLS} \frac{\Delta_0}{\Delta_1} m^3 s^{-1}. \quad (41)$$

For τ_ϕ to be on the order of 1 ns, the two-level defect density should be

$$n_{TLS} \sim \frac{\Delta_1}{\Delta_0} (10^{24} - 10^{25}) m^{-3}. \quad (42)$$

Now let us estimate n_{TLS} for a typical single-crystal system. Single-crystal silicon structures have been studied in this context in the temperature range of interest, below 1 K down to 5 mK. Both acoustic dissipation and heat capacity measurements on silicon resonators by Kleiman, Agnolet, and Bishop³⁷ (see the corresponding estimates by Phillips³⁸ and Keyes³⁹) find that the TLS density, $n_{TLS} \sim 10^{23} m^{-3}$, with an estimated value of $\Delta_1/\Delta_0 \sim 100$. Now using the same value for Δ_1/Δ_0 in the expression (42), the order-of-magnitude estimate of TLS density is found to be $n_{TLS} \sim 10^{26} - 10^{27} m^{-3}$. The required density needs to be at least three orders of magnitude higher than the typical concentration in the silicon structures to result in a TLS-induced dephasing time $\tau_\phi \sim 1$ ns. This is an unreasonably large number, even for the typical intentionally doped semiconducting structures of silicon.⁴⁰ Though, experimental studies of acoustic and thermal properties of gallium arsenide structures/heterostructures for the effects of two-level systems have not been done in the temperature range of interest for dephasing,^{21,4} recent studies on semi-insulating gallium arsenide resonators⁴¹ suggest that the typical TLS density is comparable to that in silicon.

V. CONCLUSION

We have calculated the dephasing time by assuming the presence of two-level defects inside diffusive quantum dots. The temperature dependence of $1/\tau_\phi$ is found to be roughly linear ($\sim T$) for widely distributed two-level defects in the standard tunneling model. We find that to explain the size of the experimentally observed dephasing times, we need a large number of two-level defects. This number is substantially larger than that found in glassy materials (almost by three orders of magnitude). Therefore, it is hard to believe that the electron dephasing is dominated by the intrinsic and independent two-level defects at low temperatures. We have also calculated τ_ϕ from a distribution of narrow-energy, two-level defects, and we find a regime of temperature independent τ_ϕ . However, the required number of two-level defects is too large as in the case of widely distributed TLS. The system-size dependence obtained in our calculation can be used to check the possibility of surface defects that are probably effective. Because of the large surface-to-volume ratio in quantum dots, it may be reasonable to assume that most of the defects are surface aggregated. It will be interesting to estimate \bar{P} or n_{TLS} required for the observed low-temperature charge noises of quantum dots and compare to the values from dephasing time. Unfortunately, we are not aware of any quantitative theory for the quantum dot charge noise arising from the two-level defects.

Note added. In a recent paper, Aleiner, Altshuler, and Galperin⁴² have analyzed the relevance of TLS for electron dephasing. Although they use a different approach and evaluate τ_ϕ for different systems (metals, not quantum dots), their conclusions are similar to ours—that is, a substantially large concentration of TLS, \bar{P} , much larger than the typical values in metallic glasses is required for the quantitative explanation of the saturation observed in experiments on metallic wires⁴ by two-level systems.¹⁶

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APPENDIX: DERIVATION OF EQ. (21)

For the simplicity of calculation, we suppose that the TLS is initially in the state $|-\rangle$, an eigenstate of the Hamiltonian in Eq. (11) with the eigenenergy E_- (the case with $|+\rangle$ can be calculated in a similar way). The time evolution operator $U_j(t)[U_j(t)|-\rangle = c_+(t)|+\rangle + c_-(t)|-\rangle]$ of the TLS corresponding to the electron path j can be written as

$$\begin{pmatrix} c_+(t) \\ c_-(t) \end{pmatrix} = \hat{T} \exp\left[-\frac{i}{\hbar} \int_0^t dt' \mathbf{V}_I[\mathbf{r}_j(t')]\right] \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad (\text{A1})$$

where

$$\mathbf{V}_I(\mathbf{r}_j(t)) = \begin{pmatrix} \langle + | V_{Ij}[\mathbf{r}_j(t), t] | + \rangle & \langle + | V_{Ij}[\mathbf{r}_j(t), t] | - \rangle \\ \langle - | V_{Ij}[\mathbf{r}_j(t), t] | + \rangle & \langle - | V_{Ij}[\mathbf{r}_j(t), t] | - \rangle \end{pmatrix}. \quad (\text{A2})$$

To find the corresponding time evolution of the TLS states for the time-reversed paths, $[U_{jT}(t)|-\rangle = d_+(t)|+\rangle + d_-(t)|-\rangle]$, one obtains a similar form by using $\mathbf{r}_{jT}(t) = \mathbf{r}_j(\tau_0 - t)$. Expanding $\langle - | U_{jT}^\dagger(\tau_0) U_{jT}(\tau_0) | - \rangle = c_+^*(\tau_0) d_+(\tau_0) + c_-^*(\tau_0) d_-(\tau_0)$ up to the second order in interaction \hat{V} , we get

$$\begin{aligned} & \text{Re} \langle - | U_{jT}^\dagger(\tau_0) U_{jT}(\tau_0) | - \rangle \\ &= 1 + \frac{1}{\hbar^2} \int_0^{\tau_0} dt \int_0^{\tau_0} dt' [\cos\{\Omega(t+t'-\tau_0)\} - \cos \\ & \quad \times \{\Omega(t-t')\}] \langle - | \hat{V}[\mathbf{r}_j(t)] | + \rangle \langle + | \hat{V}[\mathbf{r}_j(t')] | - \rangle, \end{aligned} \quad (\text{A3})$$

where $\hbar\Omega = E = E_+ - E_- = \sqrt{\Delta^2 + \Delta_0^2}$, and we used the relation $\hat{V}[\mathbf{r}_j(t)]^\dagger = \hat{V}[\mathbf{r}_j(t)]$ in the last equality.

Now from Eqs. (18), (A3), and (10), one can get the coherent return probability in

$$\begin{aligned} P_{\mathbf{r}}^{coh}(\tau_0) &= \sum_j |A_j(\mathbf{r}, \mathbf{r}; \tau_0)|^2 \\ &+ \frac{|\langle + | \hat{p} | - \rangle|^2}{3\hbar^2 L^{2d}} \int_0^{\tau_0} dt \int_0^{\tau_0} dt' \\ &\times [\cos\{\Omega(t+t'-\tau_0)\} - \cos\{\Omega(t-t')\}] \\ &\times \sum_{\mathbf{q}} q^2 |v_{\mathbf{q}}|^2 \sum_j |A_j(\mathbf{r}, \mathbf{r}; \tau_0)|^2 e^{i\mathbf{q} \cdot [\mathbf{r}_j(t) - \mathbf{r}_j(t')]} \end{aligned} \quad (\text{A4})$$

Here, we have omitted the subscript η in $P_{\mathbf{r}, \eta}^{coh}$ for the TLS state, because both of the initial states of the TLS $|\eta\rangle$

$= |\pm\rangle$ give rise to the same expression. $P_{\mathbf{r}}^{coh}(\tau_0)$ in Eq. (A4) is the value averaged over the TLS position. Here, we have used the disorder average over TLS $\langle\langle \exp[i\mathbf{R} \cdot (\mathbf{q} + \mathbf{q}')] \rangle\rangle = \delta_{\mathbf{q}, -\mathbf{q}'}$. The factor 3 in Eq. (A4) comes from the average over the orientation of the TLS dipoles.

In the case of $\sqrt{D\tau_0} > l$ (l is the mean free path), the sum over the classical paths that appears in Eq. (A4) can be written as a path integral using the Wiener measure.^{43,44,33} The path integral can be calculated as

$$\begin{aligned} & \sum_j |A_j(\mathbf{r}, \mathbf{r}; \tau_0)|^2 e^{i\mathbf{q} \cdot [\mathbf{r}_j(t) - \mathbf{r}_j(t')]} \\ &= \int_{\mathbf{x}(0)=\mathbf{r}}^{\mathbf{x}(\tau_0)=\mathbf{r}} D[\mathbf{x}(\tau)] \exp\left(-\frac{1}{4D} \int_0^{\tau_0} d\tau |\dot{\mathbf{x}}(\tau)|^2\right) \\ & \quad \times \exp(i\mathbf{q} \cdot [\mathbf{x}(t) - \mathbf{x}(t')]) \\ &= \frac{1}{\mathcal{V}} \sum_{|\mathbf{q}'| < \pi/l} \exp(-D|\mathbf{q}'|^2 \tau_0) e^{-D(|\mathbf{q}|^2 - 2\mathbf{q} \cdot \mathbf{q}')|t-t'|}, \end{aligned} \quad (\text{A5})$$

where we have used the boundary conditions for the quantum dot as a rectangular box with volume $\mathcal{V} = L \times L \times L$ (L or $a \ll L$ for two-dimensional dots). Several remarks are in order. Equations (A5) are valid only in diffusive regimes. The summation over the discretized momentum variables \mathbf{q} and \mathbf{q}' is understood to be limited by π/l . The contributions from ballistic regime, which are supposed to be small when $\sqrt{D\tau_0} \gg l$, are neglected in this work. The last equality in the Eq. (A5) is obtained for $\mathbf{r} = 0$.

Then, the classical return probability $P^{class}(\tau_0)$ for $\mathbf{r} = 0$ is given by

$$\begin{aligned} P^{class}(\tau_0) &= \sum_j |A_j(\mathbf{r}, \mathbf{r}; \tau_0)|^2 |_{\mathbf{r}=0} \\ &= \frac{1}{\mathcal{V}} \sum_{|\mathbf{q}| < \pi/l} \exp(-D|\mathbf{q}|^2 \tau_0). \end{aligned} \quad (\text{A6})$$

Inserting Eq. (A5) into Eq. (A4), we get the coherent part of the return probability

$$\begin{aligned} P^{coh}(\tau_0) &= \frac{1}{\mathcal{V}} \sum_{\mathbf{q}'} \exp(-D|\mathbf{q}'|^2 \tau_0) \\ &\times \left[1 + \sum_{\mathbf{q}} \frac{|\langle + | \hat{p} | - \rangle|^2 q^2 v_{\mathbf{q}}^2}{3\hbar^2 L^{2d}} \int_0^{\tau_0} dt^+ \int_{-t^+}^{t^+} dt^- \right. \\ & \quad \left. \times \{\cos \Omega(t^+ - \tau_0) - \cos \Omega t^-\} e^{-D(|\mathbf{q}|^2 - 2\mathbf{q} \cdot \mathbf{q}')|t-t'|} \right], \end{aligned} \quad (\text{A7})$$

where we used the change of variables of $t^+ = t + t'$ and $t^- = t - t'$.

We restrict to the ergodic regime $\tau_0 > \tau_D$, therefore significant contribution comes only from $\mathbf{q}' = 0$. By taking $\mathbf{q}' = 0$ and using the Fourier transform of $e^{-D|\mathbf{q}|^2|t-t'|}$ in Eq. (A7), we obtain Eq. (21).

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