Spectral scrambling in Coulomb-blockade quantum dots

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Abstract

We estimate the fluctuation width of an energy level as a function of the number of electrons added to a Coulomb-blockade quantum dot. A microscopic calculation in the limit of Koopmans' theorem predicts that the standard deviation of these fluctuations is linear in the number of added electrons, in agreement with a parametric random-matrix approach. We estimate the number of electrons it takes to scramble the spectrum completely in terms of the interaction strength, the dimensionless Thouless conductance, and the symmetry class.

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The simplest model for describing a quantum dot in the Coulomb-blockade regime [1] is the constant interaction (CI) model, in which the electrons occupy single-particle levels and the Coulomb interaction is taken as an average electrostatic energy that depends only on the number of electrons. When the single-particle dynamics in the dot are chaotic (or diffusive), one can apply random matrix theory (RMT) to describe the statistical properties of the single-particle wave functions and energies within an energy window whose width is the Thouless energy. RMT was successful in describing the conductance peak height distributions and their sensitivity to time-reversal symmetry [2,3]. However, other measured statistics, most notably the peak spacing distribution [4], indicated that it is necessary to include interaction effects beyond the simple CI model. The best way to take into account interactions while retaining a single-particle framework is the Hartree-Fock (HF) approximation, which has been used to explain some of the observed features of the peak spacing statistics [5–7].

The HF single-particle wave functions and energies are calculated self-consistently and therefore can change as electrons are added to the dot. In the statistical regime (i.e., in a chaotic or diffusive dot), this phenomenon is called scrambling. Scrambling was observed in the decay of correlations between the m-th excited state in the dot and the ground-state of a dot with an additional m electrons [8], and indirectly through the saturation of the peak-to-peak correlations as a function of temperature [9,10]. Numerical evidence of scrambling was found in the HF calculations (see, e.g., in Ref. [6]). A phenomenological way to describe scrambling is to consider a discrete set of Hamiltonians (corresponding to the different number of electrons in the dot) that are random but have the correct symmetries. Such a set is known as a discrete Gaussian process (GP) [11], and can be embedded in a continuous GP, i.e., random matrices that depend on a continuous parameter. This approach leads to a nearly Gaussian peak spacing distribution [12], and explains the saturation of the number of correlated peaks versus temperature [10]. While the parametric approach is appealing in its simplicity, it is not clear how well it describes features obtained in a microscopic approach of adding electrons into the dot. For example, in the parametric approach, the addition of m

electrons into the dot results in a fluctuation width of a typical level that is proportional to m. One can also evaluate the fluctuation width of a typical energy level in the microscopic approach through the fluctuations of the diagonal interaction matrix elements (in the limit where the single-particle wave functions do not change with the addition of electrons). If the addition of electrons is a random process, we expect the fluctuation width of a level to behave like $\sqrt{m}$. We shall show, however, that since the correlations among the diagonal interaction matrix elements are comparable to their variance, the standard deviation of the fluctuations is linear in m (for $1 \ll m \ll g$, where g is the Thouless conductance), in overall agreement with parametric RMT. The main results of the microscopic approach are summarized in Eqs. (10), (14) and (15). Extrapolating our results to larger values of m, we also estimate the dependence of the number of added electrons required for complete scrambling on g and the interaction strength (see Eq. (16)).

Scrambling implies variation of both eigenstates and energy levels. However, in this work we are concerned with the limit where the wave functions do not change and only the energy levels scramble. In the parametric approach this is valid when the change in the parameter upon the addition of m electrons is small compared to the mean (parametric) distance between avoided crossings. In the microscopic approach the appropriate limit is known as Koopmans' theorem [13], where the variation of an energy level upon the addition of an electron is just a corresponding diagonal interaction matrix element.

We first discuss the parametric approach. The variation of the single-particle energies and eigenfunctions (e.g., in a mean-field approximation) with the addition of electrons into the dot is described by a parametric variation of the Hamiltonian [14]. We denote by $H(x_{\mathcal{N}})$ the effective single-particle Hamiltonian of the dot with $\mathcal{N}$ electrons. We assume that the dot is either diffusive or ballistic with chaotic dynamics, and that the statistical properties of $H(x_{\mathcal{N}})$ are not modified by the interactions. Restricting ourselves to the universal regime, i.e., to $\sim g$ levels in the vicinity of the Fermi energy [15], we assume that $H(x_{\mathcal{N}})$ belongs to one of the Gaussian ensemble of random matrices whose symmetry class β is independent of $x_{\mathcal{N}}$. The sequence of Hamiltonians $H(x_{\mathcal{N}})$ forms a discrete GP that can be embedded in

a continuous GP $H(x)$ [19]. A simple GP is given by [11,20]

$$H(x) = \cos x H_1 + \sin x H_2 \ , \tag{1}$$

where H_1 and H_2 are $N \times N$ uncorrelated random matrices chosen from the Gaussian ensemble of symmetry class β: $P(H) \propto e^{-\frac{\beta}{2a^2}\mathrm{Tr}H^2}$. We choose $a = (2/N)^{1/2}\zeta$ so that the average level density (a semicircle) has a constant band width of $2a\sqrt{2N} = 4\zeta$, and the mean level spacing in the middle of the spectrum is $\Delta = \pi a/\sqrt{2N} = \pi\zeta/N$. The average distance between avoided crossings is given by the inverse of the rms level velocity $\delta x_c = \Delta[\overline{(\partial\epsilon_\alpha/\partial x)^2}]^{-1/2} = \pi(\beta/2N)^{1/2}$, where ϵ_α is an energy level. This distance is larger for the GUE by a factor of $\sqrt{2}$ compared with the GOE. Two-point parametric correlators become universal when the parameter x is measured in units of δx_c; i.e., as a function of a scaled parameter $\bar{x} \equiv x/\delta x_c$.

The energy levels scramble as the parameter $\bar{x}$ changes. A change of $\delta\bar{x}$ leads to a corresponding change in the energy of a single-particle level α: $\delta\epsilon_\alpha = \epsilon_\alpha(\bar{x} + \delta\bar{x}) - \epsilon_\alpha(\bar{x})$. The variance of this parametric fluctuation of a level can be estimated in the limit $\delta\bar{x} \ll 1$ using first order perturbation theory in $\delta\bar{x}$, i.e., ignoring the change of the single-particle wave function as $\bar{x} \to \bar{x} + \delta\bar{x}$:

$$\sigma^2(\delta\epsilon_\alpha) = \Delta^2(\delta\bar{x})^2 \ . \tag{2}$$

In the parametric approach it is assumed that $\bar{x}$ changes by $\delta\bar{x}_1$ upon the addition of one electron into the dot (independently of the number of electrons $\mathcal{N}$). Thus for the addition of m electrons $\delta\bar{x}_m = m\delta\bar{x}_1$, and as long as $\delta\bar{x}_m \ll 1$, we can use (2) to estimate the variance of the change in the energy level α when m electrons are added

$$\sigma(\delta\epsilon_\alpha^{(m)}) = \Delta m\delta\bar{x}_1 = m\sigma(\delta\epsilon_\alpha^{(1)}) \ , \tag{3}$$

where $\delta\epsilon_\alpha^{(m)} \equiv \epsilon_\alpha^{(\mathcal{N}+m)} - \epsilon_\alpha^{(\mathcal{N})}$ ($\epsilon_\alpha^{(\mathcal{N}+m)}$ is the energy of level α in a dot with $\mathcal{N}+m$ electrons).

To relate the parametric approach to a microscopic mean-field approach, we describe the effect that adding one or a few electrons has on a particular single-particle HF level. The Hamiltonian of the dot is

$$H = \sum_{ij} h^{(0)}_{ij} a^\dagger_i a_j + \frac{1}{4} \sum_{ijkl} v^A_{ijkl} a^\dagger_i a^\dagger_j a_l a_k \ , \tag{4}$$

where $h^{(0)}$ is the one-body part (accounting for the disorder or the chaotic confining potential) and $v^A_{ij;kl} \equiv \langle ij|v|kl\rangle - \langle ij|v|lk\rangle$ are the antisymmetrized matrix elements of the two-body interaction. The single-particle HF Hamiltonian for $\mathcal{N}$ electrons, $h^{(\mathcal{N})}_{\alpha\gamma} = \epsilon^{(0)}_{\alpha\gamma} + \sum_{\delta=1}^{\mathcal{N}} v^A_{\alpha\delta;\gamma\delta}$, and its eigenvalues $\epsilon^{(\mathcal{N})}_\alpha$ are determined self-consistently by the HF equations $h^{(\mathcal{N})}_{\alpha\gamma} = \epsilon^{(\mathcal{N})}_\alpha \delta_{\alpha\gamma}$. In the HF basis, the *diagonal* part of the Hamiltonian (4) is

$$H_{\rm diagonal} = \sum_\alpha \epsilon^{(0)}_{\alpha\alpha} \hat{n}_\alpha + \frac{1}{2} \sum_{\alpha,\gamma} v^A_{\alpha\gamma} \hat{n}_\alpha \hat{n}_\gamma \tag{5}$$

where $\hat{n}_\alpha$ is the number operator for single-particle state α, and we have denoted $v^A_{\alpha\gamma} \equiv v^A_{\alpha\gamma;\alpha\gamma}$.

In the limit of Koopmans' theorem, the single-particle wave functions do not change when an electron is added to the dot. Consequently, the change in the single-particle energy ϵ_α when an electron is added to the $\mathcal{N}+1$-st level is given by a diagonal interaction matrix element

$$\delta\epsilon^{(1)}_\alpha \equiv \epsilon^{(\mathcal{N}+1)}_\alpha - \epsilon^{(\mathcal{N})}_\alpha \approx v^A_{\alpha\mathcal{N}+1} \ . \tag{6}$$

Assuming the HF wave functions satisfy similar statistics as in the non-interacting case, we can estimate the variance of the matrix element in (6). As long as the relevant single-particle levels are in the universal regime, the variance of a diagonal matrix element $v_{\alpha\gamma}$ is independent of the particular orbits α and γ. The two-body interaction used in the HF approximation is often an effective interaction, e.g., an RPA screened interaction [17]. Alternatively, one can employ a short-range dressed interaction [18] $v(\boldsymbol{r}-\boldsymbol{r}') = \lambda\Delta V \delta(\boldsymbol{r}-\boldsymbol{r}')$, where λ is the interaction strength and V is the system's volume. A diagonal interaction matrix element is then given by $v_{\alpha\gamma} = \lambda\Delta V \int d\boldsymbol{r} \psi^*_\alpha(\boldsymbol{r})\psi^*_\gamma(\boldsymbol{r})\psi_\alpha(\boldsymbol{r})\psi_\gamma(\boldsymbol{r})$, and its variance

$$\begin{aligned}\sigma^2(v_{\alpha\gamma}) = \lambda^2\Delta^2 V^2 \int d\boldsymbol{r}_1 \int d\boldsymbol{r}_2 \Big[&\Big\langle \psi^*_\alpha(\boldsymbol{r}_1)\psi^*_\gamma(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_2)\psi_\gamma(\boldsymbol{r}_2)\psi^*_\alpha(\boldsymbol{r}_2)\psi^*_\gamma(\boldsymbol{r}_2)\Big\rangle \\ &- \Big\langle \psi^*_\alpha(\boldsymbol{r}_1)\psi^*_\gamma(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_1)\Big\rangle \Big\langle \psi_\alpha(\boldsymbol{r}_2)\psi_\gamma(\boldsymbol{r}_2)\psi^*_\alpha(\boldsymbol{r}_2)\psi^*_\gamma(\boldsymbol{r}_2)\Big\rangle \Big]\end{aligned} \tag{7}$$

Only the connected part of the ensemble average over the product of eight wave functions contributes to the r.h.s. of (7). In the following we restrict our calculations to the GUE symmetry. All possible pairwise and quadruplet-wise contractions in (7) should be taken into account, and they can be regrouped to give [21]

$$\sigma^2(v_{\alpha\gamma}) = \lambda^2\Delta^2V^2\int d\boldsymbol{r}_1\int d\boldsymbol{r}_2\left\{\left[\langle\psi^*_\alpha(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_2)\psi^*_\alpha(\boldsymbol{r}_2)\rangle\left\langle\psi^*_\gamma(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_2)\psi^*_\gamma(\boldsymbol{r}_2)\right\rangle - \frac{1}{V^4}\right]\right.$$
$$+\left[\left\langle\psi^*_\alpha(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_2)\psi^*_\gamma(\boldsymbol{r}_2)\right\rangle\left\langle\psi^*_\gamma(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_2)\psi^*_\alpha(\boldsymbol{r}_2)\right\rangle - \frac{1}{V^4}\right]$$
$$\left.+\left\langle\psi^*_\alpha(\boldsymbol{r}_1)\psi_\alpha(\boldsymbol{r}_2)\psi_\gamma(\boldsymbol{r}_1)\psi^*_\gamma(\boldsymbol{r}_2)\right\rangle\left\langle\psi^*_\gamma(\boldsymbol{r}_1)\psi_\gamma(\boldsymbol{r}_2)\psi_\alpha(\boldsymbol{r}_1)\psi^*_\alpha(\boldsymbol{r}_2)\right\rangle\right\}\ , \tag{8}$$

where the $1/V^4$ factors are subtracted to avoid double counting of pairwise contractions.

Wave-function correlations appearing in Eq. (8) were calculated in Refs. [22,24]. Including contributions of order $1/g^2$, we have

$$\sigma^2(v_{\alpha\gamma}) = \frac{\lambda^2\Delta^2}{V^2}\int d\boldsymbol{r}_1\int d\boldsymbol{r}_2\left\{\left[1+k_d(r)(1+\Pi(\boldsymbol{r}_1,\boldsymbol{r}_1))+\Pi(\boldsymbol{r}_1,\boldsymbol{r}_2)+b\Pi^2(\boldsymbol{r}_1,\boldsymbol{r}_2)\right]^2-1\right.$$
$$+\left[1+k_d(r)\Pi(\boldsymbol{r}_1,\boldsymbol{r}_1)+c\Pi^2(\boldsymbol{r}_1,\boldsymbol{r}_2)\right]^2-1$$
$$\left.+\left[k_d(r)+\Pi(\boldsymbol{r}_1,\boldsymbol{r}_2)\right]^2\right\}\ , \tag{9}$$

where b, c are coefficients of order 1 and $\boldsymbol{r} = \boldsymbol{r}_1 - \boldsymbol{r}_2$. In Ref. [24] it was found that $c = 1/2$. The function $k_d(r) \equiv (\pi\nu)^2\langle \mathrm{Im}G^R(\boldsymbol{r})\rangle^2$ (where G^R is the retarded Green function and ν is the electron density of states per unit volume) describes the short-range correlations. In 2D $k_d(r) = \exp(-r/\ell)J_0(k_Fr)$ (where ℓ is the mean free path and J_0 is the Bessel function of order zero), while in 3D $k_d(r) = \exp(-r/\ell)(\sin k_Fr/k_Fr)^2$. $\Pi(\boldsymbol{r}_1,\boldsymbol{r}_2) = (\pi\nu)^{-1}\sum_{\boldsymbol{Q}\neq 0}\phi_{\boldsymbol{Q}}(\boldsymbol{r}_1)\phi_{\boldsymbol{Q}}(\boldsymbol{r}_2)/D\boldsymbol{Q}^2$ is the long-range diffuson contribution where $\phi_{\boldsymbol{Q}}$ is the eigenfunction of the diffusion operator corresponding to the eigenvalue $D\boldsymbol{Q}^2$. The main contributions to (9) arise from the long range terms $\sim\Pi^2$ (note that $\int d\boldsymbol{r}_1\Pi(\boldsymbol{r}_1,\boldsymbol{r}_2) = 0$ and does not contribute). We find

$$\sigma^2(v_{\alpha\gamma}) = 2(b+c+1)\kappa\left(\lambda\Delta/g\right)^2\ , \tag{10}$$

where $g = 2\pi\nu DL^{d-2}$ is the dimensionless Thouless conductance. The quantity $\kappa = \frac{1}{4\pi^4}\sum_{\boldsymbol{n}\neq 0}1/\boldsymbol{n}^4$, where the summation is over $\boldsymbol{n} = (n_1,\ldots,n_d)$ with n_i non-negative integers, and only the term $\boldsymbol{n} = (0,\ldots,0)$ is excluded. We remark that the contributions

leading to the terms with b and c in Eq. (10) were ignored in deriving the variance of a diagonal matrix element in Ref. [24].

Comparing the variance (2) of the change in an energy level in the parametric approach with the corresponding variance (10) in the microscopic approach when a single electron is added (see Eq. (6)), we obtain the scrambling parameter

$$\delta\bar{x}_1 = [2(b+c+1)\kappa]^{1/2}\frac{\lambda}{g} \ . \tag{11}$$

If the symmetry class is taken into account then $\delta\bar{x}_1 \sim \beta^{-1}\lambda/g$. The addition of an electron to the finite dot can lead to excess negative charge on the boundaries of the dot [17]. Repeating the calculation that led to Eqs. (10) and (11) for the one-body potential generated by this surface charge, we find $\delta\bar{x}_1 \sim \beta^{-1/2}\lambda/g^{1/2}$ [10].

We now consider the addition of m electrons into the dot. Extending Koopmans' limit to m electrons, i.e., assuming the single-particle wave functions do not change with the addition of m electrons, we can express the change in a single-particle level α as

$$\delta\epsilon_\alpha^{(m)} \equiv \epsilon_\alpha^{(\mathcal{N}+m)} - \epsilon_\alpha^{(\mathcal{N})} \approx \sum_{i=1}^{m} v_{\alpha\alpha+i}^A \ . \tag{12}$$

The variance of (12) is given by $\sigma^2(\delta\epsilon_\alpha^{(m)}) = m\sigma^2(v_{\alpha\gamma}) + m(m-1)(\overline{v_{\alpha\gamma}v_{\alpha\delta}} - \overline{v_{\alpha\gamma}}\ \overline{v_{\alpha\delta}})$, where we have assumed all single-particle wave functions $\alpha,\ldots,\alpha+m$ to be in the universal regime, i.e., the variances and covariances are independent of the particular wave functions. The covariance of $v_{\alpha\gamma}$ and $v_{\alpha\delta}$ can be calculated similarly to the variance calculation. We obtain

$$\begin{aligned}\overline{v_{\alpha\gamma}v_{\alpha\delta}} - \overline{v_{\alpha\gamma}}\ \overline{v_{\alpha\delta}} = \frac{\lambda^2\Delta^2}{V^2}\int d\boldsymbol{r}_1 \int d\boldsymbol{r}_2 \Big\{ \Big[1 + k_d(r)(1+\Pi(\boldsymbol{r}_1,\boldsymbol{r}_1)) + \Pi(\boldsymbol{r}_1,\boldsymbol{r}_2) + b\Pi^2(\boldsymbol{r}_1,\boldsymbol{r}_2)\Big] \\ \times \Big[1 + k_d(r)\Pi(\boldsymbol{r}_1,\boldsymbol{r}_1) + c\Pi^2(\boldsymbol{r}_1,\boldsymbol{r}_2)\Big] - 1 \\ + \Big[1 + k_d(r)\Pi(\boldsymbol{r}_1,\boldsymbol{r}_1) + c\Pi^2(\boldsymbol{r}_1,\boldsymbol{r}_2)\Big]^2 - 1\Big\} \ ,\end{aligned} \tag{13}$$

and to leading order

$$\overline{v_{\alpha\gamma}v_{\alpha\delta}} - \overline{v_{\alpha\gamma}}\ \overline{v_{\alpha\delta}} = (b+3c)\kappa\,(\lambda\Delta/g)^2 \ . \tag{14}$$

Expressions (10) and (14) can also be derived diagrammatically. Fig. 1 shows five diagrams that contribute to order Π^2. Since $c = 1/2$, we have the relation $\sigma^2(v_{\alpha\gamma}) = 2(\overline{v_{\alpha\gamma}v_{\alpha\delta}} - \overline{v_{\alpha\gamma}}\ \overline{v_{\alpha\delta}})$. Using (12) we find

$$\sigma^2(\delta\epsilon_\alpha^{(m)}) = \frac{1}{2}m(m+1)\sigma^2(\delta\epsilon_\alpha^{(1)}) \; . \tag{15}$$

Thus, for $m \gg 1$, the standard deviation of the change in the single-particle energy is linear in the number of added electrons m, in agreement with result (2) in the parametric approach. In particular we find that the statistical fluctuations accumulated by sequentially adding electrons into the dot are not added independently.

A complete scrambling of the spectrum corresponds to a parametric change of one avoided crossing, i.e., $\delta\bar{x}_m \sim 1$. Using (11), we can estimate the number of added electrons m_c needed to completely scramble the spectrum

$$m_c \sim \frac{1}{\delta\bar{x}_1} \sim \beta\frac{g}{\lambda} \; . \tag{16}$$

In the presence of surface charge (see discussion after Eq. (11)) we obtain instead of (16)

$$m_c \sim \frac{(\beta g)^{1/2}}{\lambda} \; . \tag{17}$$

We note that since we have neglected wave-function scrambling due to off-diagonal interaction matrix elements $v_{\alpha,\beta;\gamma,\delta}$, our estimates for m_c in Eqs. (16) and (17) should only be regarded as upper bounds.

In a recent experiment [25] the fluctuation width $\sigma_p(m) \equiv \sigma(V_{\mathcal{N}+m} - V_{\mathcal{N}})$, where $V_{\mathcal{N}}$ is the position in gate voltage for the $\mathcal{N}$-th peak, was measured as a function of the separation m in peak number. In Koopmans' limit $V_{\mathcal{N}+m} - V_{\mathcal{N}} \approx \epsilon_{\mathcal{N}+m}^{(\mathcal{N}+m)} - \epsilon_{\mathcal{N}}^{(\mathcal{N})} = \Delta_m^{(\mathcal{N}+m)} + \delta\epsilon_{\mathcal{N}}^{(m)}$, where $\Delta_m^{(\mathcal{N}+m)} \equiv \epsilon_{\mathcal{N}+m}^{(\mathcal{N}+m)} - \epsilon_{\mathcal{N}}^{(\mathcal{N}+m)}$ is the distance between levels separated by m consecutive spacings in a dot with a fixed number of electrons $(\mathcal{N}+m)$. The latter quantity $\Delta_m^{(\mathcal{N}+m)}$ is unrelated to scrambling and is completely determined (for $m < g$) from RMT. For $1 \ll m < g$, $\sigma(\Delta_m^{(\mathcal{N}+m)}) \propto \Delta \ln m$. In principle one can extract $\sigma(\delta\epsilon_{\mathcal{N}}^{(m)})$ from the measured $\sigma_p(m)$. In practice this is difficult to do with the available experimental statistics since $\sigma(\delta\epsilon_{\mathcal{N}}^{(m)}) \ll \sigma(\Delta_m^{(\mathcal{N}+m)})$ for $m \ll g$ (in the experiment $g \sim 50$), and $\sigma_p(m)$ is dominated by the $\ln m$ term of RMT. Nevertheless, $\sigma_p(m)$, when plotted versus $\ln m$, appears to follow a straight-line behavior with a small concave deviation (for $m \lesssim 7$) [26], in qualitative agreement with an additional contribution due to scrambling.

In conclusion, we have studied spectral scrambling when several electrons are added to a Coulomb-blockade quantum dot, and shown a correspondence between the microscopic and parametric approaches. Our analysis was carried out within the framework of the HF-Koopmans picture and was limited to energy fluctuations. Accounting for the scrambling of the wave functions microscopically is an outstanding problem.

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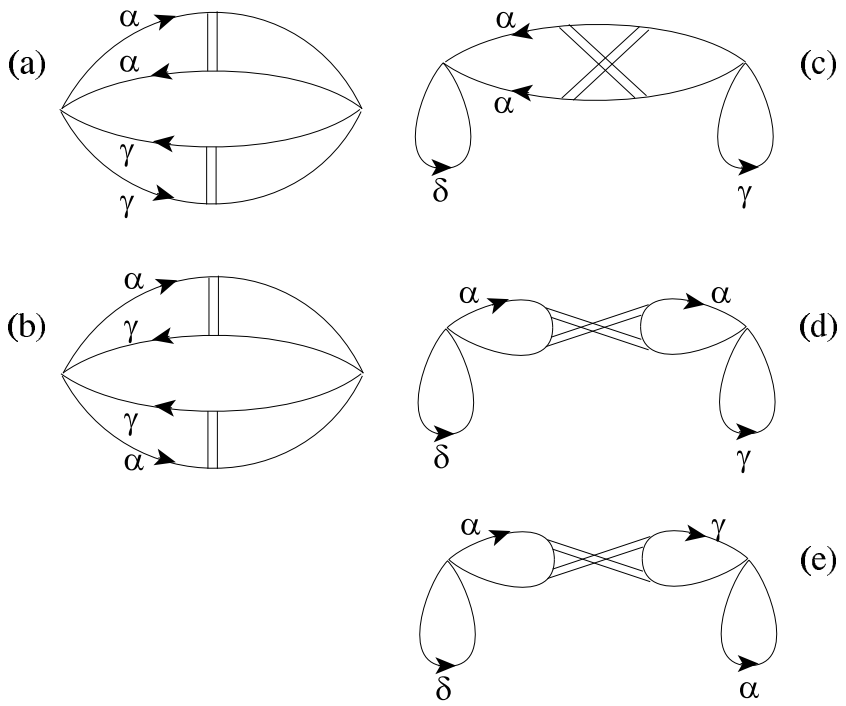

FIG. 1. Diagrams to order Π^2 in the calculation of the variance (10) and covariance (14). In diagrams (c) – (e) we take $\beta = \gamma$ when calculating (10). Diagrams (a) and (b) do not contribute to (14). Diagrams (c) and (d) contribute to the constant b, while diagram (e) – to the constant c.