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# Mixed-State Density Operator in a Nonlinear Quantum System

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## Abstract

We consider the simplest case of two nonlinear quantum eigenstates whose overlap integral defined by their inner product is non-zero. We propose to regard this system as a mixed-state quantum system obtained by statistically combining the two nonlinear eigenstates with appropriate normalized probabilities. Since this description in terms of the density operator is unambiguous only if the quantum states of the mixture are orthonormal, we use Löwdin's change of basis to symmetrically orthonormalize the original pair of non-orthogonal eigenstates and to define the corresponding density matrix. We show by simple examples that this mixed-state description is in good agreement with experimental and/or numerical results.

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Nonlinear quantum theory in its simplest form concerns N-particle interacting systems whose mean-field description allows a single-particle approximation at the price of losing the fundamental linear structure of the theory. Perhaps the earliest such attempt is the Hartree-Fock model where the resulting nonlinear Schrödinger eigenstates are as a rule not orthogonal [1]. Actually, as early as in the 30's, it became clear that the overlap integrals describing non-orthogonality, instead of being negligible, were quantities of essential importance for the understanding of the physical properties of solids. Pauling for instance [2] as well as Slater [3] developed the principle of maximum overlap, saying that the strength of a chemical bond increases with the overlap between the orbitals involved, measured essentially by the overlap integrals. As for Inglis [4]: he coined the word "nonorthogonality catastrophe". In electron-transfer models, nonvanishing overlap integrals yield effective Hamiltonians which give significantly better estimates for the eigenenergies of the system than when these integrals are neglected [5]. In the present case —and contrary to the above ones—, non-orthogonality is not just due to the choice of appropriate basis. It is a direct consequence of a much deeper change in the quantum theory itself, namely, nonlinearity [6]. A celebrated such example is the nonlinear Schrödinger —or Gross-Pitaevskii (GP)— equation for the mean-field quantum description of a Bose-Einstein condensate (BEC). It was believed that, although the corresponding nonlinear eigenstates are indeed not orthogonal, the effect of their overlap integrals remains negligible [7]. While indeed numerically small, it may yield interesting non-trivial interference effects [8].

Other fundamentally nonlinear quantum systems have been considered, for instance in the pioneering attempts to explain superconductivity. Prior to the discovery of the microscopic BCS theory of superconductivity as a Bose condensation of electrons paired by microscopic electron-lattice interactions [9], tentative links of superconductivity with the interaction between electrons and lattice vibrations have been explored [10] [11]. Most interesting in the scope of the present work is Schafroth's early suggestion that charge-carrying bosons in a metal at low temperature actually constitute a gas of bound two-electron states [12, 13] which can then be described by a self-consistent normalized stationary Schrödinger-Poisson (SP) nonlinear differential system [14]. At that time (1955) where performant numerical simulations were not available, Schafroth just wanted to emphasize the role of long-range Coulomb interactions between the charged bosons by use of mostly qualitative arguments. Therefore he proposed a one-dimensional (1D) SP model that considers the particles, whose quantum state is defined by the stationary Schrödinger equation, as moving in an effective potential related by Poisson's equation to the local boson charge density plus an uniform potential modelling both the charge density of the background (in order to make the whole system electrically neutral) and the non-condensed particles. Due to the assumed Poisson geometry, this additional uniform potential is but the second derivative of an equivalent external harmonic confining potential whose frequency is simply the plasma frequency of the electrons. Consequently, this 1955 SP model was probably the first *ab-initio* nonlinear quantum attempt to describe by use of a self-consistent differential system the competition that occurs in a charged boson gas between external confinement and long-range internal particle-particle Coulomb interaction [15].

With the experimental development of BEC's as well as quantum dots, the theoretical interest of this competition has all the more increased as nonlinear differential systems are now routinely solved by precise numerical simulations. Therefore the quantum nature of their eigenstates can now be numerically explored and the physical consequences of their nonlinearity as well as their non-orthogonality addressed. Non-orthogonal eigenstates due

to system-environment couplings lead e.g. to a plethora of intriguing non-Hermitian phenomena which can be described either by a duality between non-Hermitian models in flat spaces and their Hermitian counterparts in curved spaces [16] or by linear quantum electrodynamics directly deduced from the nonlinear SP equation [17]. Special emphasis is also put on strong nonlinearity in such systems which may yield their transition to a Thomas-Fermi classical state [18] or their description by a stable algorithm in the difficult case where the density of states varies rapidly with energy [19].

We consider in the present letter the simplest case of two nonlinear eigenstates  $|i\rangle$  and  $|j\rangle$  defined by their respective eigenvalues —or chemical potentials—  $\mu_i$  and  $\mu_j \neq \mu_i$ . How can the non-zero probability amplitude  $\langle i|j\rangle$  —defined by its overlap integral  $S_{ij} = \int u_i^* u_j d^3r \neq 0$  of their respective wave functions  $u_{i,j}(\mathbf{x})$ — be physically interpreted (in order to avoid any confusion, we use parenthesis for the nonlinear eigenstates, e.g.  $|a\rangle$ , and, as usually, kets for standard linear orthonormal eigenstates : e.g.  $|b\rangle$ )? We propose a tentative answer in terms of the well-known quantum concept of mixed-state quantum system obtained by statistically combining the nonlinear eigenstates  $\{|i\rangle\}$  with appropriate normalized probabilities. However, it is known that its description in terms of the density operator  $\rho$  is unambiguous only if the quantum states of the mixture are orthonormal. Then the statistical weights are the eigenvalues of the corresponding density matrix [20]. Therefore we make use of Löwdin's change of basis which symmetrically orthonormalizes an original non-orthogonal set of eigenstates [21] —e.g. modeling electron transport through a molecule without Green's functions or equivalent scattering theory methods [22]— and we build-up the set of Löwdin orthonormal states  $|j\rangle = \sum_i [\mathbf{S}^{-1/2}]_{j,i} |i\rangle$  where  $\mathbf{S}$  is the overlap matrix. We define the mixed state by the density matrix  $\rho \sim \{S_{i,j}\}$ , once this later is appropriately diagonalized in the new basis  $|j\rangle$ . The corresponding eigenvalues  $p_j > 0$  are the normalized statistical weights:  $\sum_j p_j = 1$  and  $\rho = \sum_j p_j |j\rangle \langle j|$ . Then we use these results to define an observable of the nonlinear quantum system, namely here the radial extend  $\langle r^2 \rangle = Tr\{\rho[\mathbf{r}^2]\}$  of a 2D axisymmetric system where  $\rho$  and  $[\mathbf{r}^2]$  are respectively the density matrix and the matrix  $r_{i,j}^2 = \langle j|\mathbf{r}^2|i\rangle$  in the nonlinear eigenbasis  $\{|i\rangle\}$ .

The above procedure is straightforward for several discrete nonlinear eigenstates. For the sake of clarity, we illustrate it by use of the simplest case, namely, a two-level nonlinear system which both is easily analytically tractable and displays the main properties of the model. Moreover, it is experimentally sound: e.g. the "s" states of two-electron quantum-dot helium [6] [23].

To be specific, , assume that the wave functions  $u_{i,j}(\mathbf{x})$  of the nonlinear normalized eigenstates  $|i\rangle$  and  $|j\rangle$  corresponding to the respective nonlinear eigenvalues  $\mu_i$  and  $\mu_j$  are real-valued. Define the eigenstate non-orthogonality  $A = \langle i|j\rangle = \langle j|i\rangle = \int u_i u_j d^3r = S_{ij} = S_{ji}$  where the overlap matrix  $\mathbf{S}$  is :

$$\mathbf{S} = \begin{pmatrix} 1 & A \\ A & 1 \end{pmatrix} \quad \longrightarrow \quad \mathbf{S}^{-1/2} = \frac{\pm 1}{\sqrt{1-A^2}} \begin{pmatrix} \cos \theta & -\sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix} , \quad (1)$$

where  $A = \sin 2\theta$ . Löwdin's symmetrically-orthonormalized states are [21]:

$$\begin{pmatrix} |i\rangle \\ |j\rangle \end{pmatrix} = \mathbf{S}^{-1/2} \begin{pmatrix} |i\rangle \\ |j\rangle \end{pmatrix} . \quad (2)$$

Note that, due to nonlinearity, these states are not *eigenstates* —nor even solutions— of the nonlinear differential quantum system (say, SP or GP for instance). They will serve as

intermediate tools in order to unambiguously define the 2x2 density matrix  $\rho$  in the basis of the two original nonlinear normalized eigenstates  $|i\rangle$  and  $|j\rangle$ . The spectral decomposition of the density matrix  $\rho$  in Löwdin's orthonormal basis  $|i\rangle$  and  $|j\rangle$  is given by:

$$\mathbf{\Lambda}_p = \begin{pmatrix} p_i & 0 \\ 0 & p_j \end{pmatrix} = \mathbf{S}^{1/2} \rho \mathbf{S}^{-1/2} \iff \rho = \sum_{l=i,j} p_l |l\rangle\langle l| \quad , \quad (3)$$

where the eigenvalues (or statistical weights)  $p_{i,j}$  are:

$$p_i = \frac{1}{2} \pm (\alpha - \frac{1}{2})\sqrt{1 - A^2} \quad ; \quad p_j = \frac{1}{2} \mp (\alpha - \frac{1}{2})\sqrt{1 - A^2} \quad . \quad (4)$$

The  $\pm$  sign is redundant: it defines two actually equivalent solutions that only depend on eigenstate labelling. Therefore we adopt in the following the positive sign. The normalization condition  $\sum_{l=i,j} p_l = 1$  yields the following solution in the nonlinear basis  $|i\rangle$  of the spectral problem (3):

$$\rho = \begin{pmatrix} \alpha & A(\alpha - \frac{1}{2}) \\ -A(\alpha - \frac{1}{2}) & 1 - \alpha \end{pmatrix} \quad . \quad (5)$$

Parameter  $\alpha$ , which defines —together with non-orthogonality parameter  $A = \langle i|j\rangle$ — the populations of Löwdin's orthogonal states  $|i\rangle$  or  $|j\rangle$  in accordance with Eq. (4), is the only adjustable degree of freedom in our nonlinear quantum model  $|i, j\rangle$  in order to fit with experimental values. Indeed consider any Hermitian operator  $\mathbf{Q}$  whose matrix in the real-valued nonlinear basis  $|i, j\rangle$  is symmetric:  $Q_{ij} = \int u_i \mathbf{Q} u_j d^3r = \int u_j \mathbf{Q} u_i d^3r = Q_{ji}$ . Therefore:

$$\langle Q \rangle = Tr\{\rho \mathbf{Q}\} = \alpha Q_{ii} + (1 - \alpha) Q_{jj} \iff \alpha = \frac{\langle Q \rangle - Q_{jj}}{Q_{ii} - Q_{jj}} \quad . \quad (6)$$

As an illustration, assume that  $\mathbf{Q}$  is the Hamiltonian  $\mathbf{H}$  which yields in the nonlinear basis  $|i, j\rangle$ :  $H_{ii} = \mu_i$ ;  $H_{ij} = H_{ji} = 0$ ;  $H_{jj} = \mu_j$ . Then the energy reads  $E = Tr\{\rho \mathbf{H}\} = \alpha \mu_i + (1 - \alpha) \mu_j$  or  $\alpha = (E - \mu_j) / (\mu_i - \mu_j)$  in accordance with Eqs (6). Consider the quantization of the two-electron Coulomb system [15]: the ground state nonlinear eigenvalue (say,  $\mu_i$ ) is by less than 1% lower than the corresponding experimental value  $E$ . Therefore  $\alpha \sim 0.99$ . Specifically, the  $1s^2$  carbon ion  $C^{4+}$  yields [15]:  $\mu_i = -0.40059 \text{ a.u.}/Z^2$  and  $\mu_j = -0.1007 \text{ a.u.}/Z^2$  (with  $Z = 6$ ) while the corresponding experimental value is  $E = -0.40023 \text{ a.u.}/Z^2$ . Therefore  $\alpha \sim 0.9988$  and the corresponding density matrix (5) reads:

$$\rho = \begin{pmatrix} 0.9988 & -0.0249 \\ 0.0249 & 0.0012 \end{pmatrix} \quad . \quad (7)$$

since  $A \sim -0.05$  [15]. Another example concerns the time-dependent axisymmetric two-state SP nonlinear system whose ground state  $u_i(x, y)$  is initially perturbed ("kicked") by a sudden modification of the bottom of its external parabolic confining potential. We consider the case  $A = -0.132$  (a realistic value for GaAs since it corresponds to the harmonic confinement  $\hbar\omega = 3.37 \text{ meV}$  [6]). By numerically projecting the resulting time-dependent wave function  $\Psi(x, y, t)$  onto ground state  $u_i(x, y)$ , then identifying  $p_i$  with the mean value 0.974 of this oscillating component, and making use of:

$$\alpha = \frac{1}{2} + \frac{1}{\sqrt{1 - A^2}} (p_i - \frac{1}{2}) \quad , \quad (8)$$

deduced from Eq. (4), we obtain  $\alpha = 0.978$ . Choosing  $\mathbf{Q} = [\mathbf{X}^2]$  where  $X^2$  is the 2D extend of the system in the above dimensionless units, we have  $X_{ii}^2 = 3.64$  and  $X_{jj}^2 = 7.38$ . Therefore, by use of Eqs. (6), we obtain  $\langle r^2 \rangle = Tr\{\rho[\mathbf{r}^2]\} = 7.38 - 3.74\alpha = 3.72$  in appropriate dimensionless units; to be compared with the actual averaged value 3.77 given by the time-dependent numerical solution of the "kicked" SP differential system [24]. Therefore our two-state stationary SP model, together with its nonlinear density matrix (4-5), provides quite acceptable mean values when compared with the corresponding averaged time-dependent description.

In conclusion, let us stress that the generalization of the above two-level mixed-state density operator description to a system where several nonlinear quantum eigenstates coexist is mathematically straightforward. The use of the density matrix, together with its unavoidable Löwdin change of basis orthonormalizing the original non-orthogonal set of eigenstates [21], matters here because it is the only physical tool to take into account the mixed-state properties of the quantum system. Indeed these latter are obtained by statistically combining the nonlinear eigenstates with appropriate normalized probabilities. These mixed-state properties can be valuable for the description of, e.g., both the charged Bose gas [15] and the GP system [25]. In the former case, the interest of the nonlinear eigenstates is that they can be regarded as unperturbed although they take into account, in addition to the particles' external parabolic confinement, the (usually quite important) long-range Coulomb interactions [26]. The mathematical consequence is their non-orthogonality yielding in particular their mixed-state properties. In the later case, nonlinear quantum eigenstates in the physical description of very-many-particle stationary BEC's are practically quasi-unavoidable. Indeed, when the particle number exceeds a few thousands, there is actually no other choice than solving the nonlinear GP equation. Actually, the ground state of  $N$  interacting bosons confined by an external potential can be directly calculated starting from its many-body Hamiltonian and using a path-integral Monte Carlo method (e.g. in order to describe the thermodynamic behaviour of many thousands of atoms interacting with a repulsive "hard-sphere" potential [27]). Although this procedure gives exact results within statistical errors, the calculation can be heavy or even impracticable for systems with much larger values of  $N$ . Therefore mean-field approaches are commonly developed for interacting systems in order to overcome the problem of solving exactly the full many-body Schrödinger equation. Apart from the convenience of avoiding heavy numerical work, they allow one to understand the behaviour of a system in terms of a set of parameters having a clear physical meaning, particularly in the case of trapped bosons [25].

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