Quantum-state tomography for spin-*l* systems

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We show that the density matrix of a spin-l system can be described entirely in terms of the measurement statistics of projective spin measurements along a minimum of $4l+1$ different spin directions. It is thus possible to represent the complete quantum statistics of any N-level system within the spherically symmetric three-dimensional space defined by the spin vector. An explicit method for reconstructing the density matrix of a spin-1 system from the measurement statistics of five nonorthogonal spin directions is presented and the generalization to spin-l systems is discussed.

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I. INTRODUCTION

As rapid progress is being made in the experimental generation of quantum states, it becomes necessary to develop efficient methods of characterizing the actual mixed state output of each new realization. In particular, various types of optical spin-1 systems have recently been generated using parametric down-conversion [1–6]. It is therefore interesting to consider the measurements necessary to properly identify the quantum states of such spin systems.

In the most general case, these states can be characterized by reconstructing the complete density matrix from a sufficiently large set of measurements, a procedure commonly referred to as quantum tomography [7–10]. For two-level systems (qubits), quantum tomography is usually realized by measuring the three orthogonal components of the Bloch vector represented by the Pauli operators. In spin-1/2 systems, the physical meaning of these components is generally clear. In particular, they represent the components of the three-dimensional Stokes vector in the commonly studied case of single-photon polarization [7,8]. In spin-l systems with higher total spin, the connection between the much larger number of density-matrix elements and the physical properties of the system is less clear. For abstract N-level systems (qudits), an expansion of the density matrix into the generators most closely related to the individual density-matrix elements has been proposed [9]. However, the physical properties corresponding to these operators are quite different from the spin components observed, e.g., in Stern-Gerlach or n-photon polarization measurements.

In particular, the recently generated n-photon polarization states are usually characterized by photon detection measurements in a pair of orthogonal polarization directions [1–4]. This corresponds to a projective measurement of one component of the three-dimensional Stokes vector, which is formally equivalent to the three-dimensional spin vector. The direction of the Stokes vector component is determined by the pair of orthogonal polarization directions detected in the measurement and can be varied by using standard birefringent elements such as quarter wave plates and half wave plates. The experimental characterization of such optical spin-l systems thus corresponds to the measurement of spin components $\hat{L}_i$ along a set of well-defined measurement directions i. It is therefore desirable to formulate quantum tomography in terms of the measurement statistics obtained in this kind of measurements.

In the following, we show how the measurement statistics obtained in measurements of spin components $\hat{L}_i$ relate to the elements of the density matrix. Based on these results, a systematic approach to the quantum tomography of spin-l systems is developed. We propose a decomposition of the density matrix into components that reflect the spherical symmetry of the spin system and correspond directly to well-defined contributions in the experimentally observable spin statistics. It is shown that measurements along a minimum of $4l+1$ spin directions are necessary to reconstruct the complete density matrix. An explicit method for reconstructing a spin-1 density matrix from the measurement probabilities along five nonorthogonal spin directions is derived and the extension of this method to general spin-l systems is discussed. Since this method can be applied equally well to small (few-level) and large (many-level) quantum systems, it also provides a useful basis for the discussion of decoherence and the transition from quantum to classical physics.

II. MEASUREMENT STATISTICS OF A SPIN-*l* SYSTEM

Each projective von Neumann measurement of a spin component $\hat{L}_i$ results in one of the $2l+1$ eigenvalues m of the quantized spin along the direction corresponding to i. By repeating the measurement a large number of times, it is possible to determine the probability distribution $p_i(m)$ of the measurement outcomes m. The information represented by this probability distribution can also be expressed in terms of averages of different powers of $\hat{L}_i$,

$$\langle (\hat{L}_i)^n \rangle = \sum_m m^n p_i(m). \tag{1}$$

The probability distribution over the $2l+1$ possible outcomes is then uniquely defined by the $2l$ averages obtained for n

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$=1$ to $n=2l$; that is, the $2l$ expectation values form a set of linearly independent parameters describing the complete measurement statistics.

Using this representation of the measurement information obtained along one spin direction, it is now possible to derive the relations between the measurement statistics along different spin directions by expressing the spin statistics of an arbitrary spin direction in terms of the three orthogonal spin components $\hat{L}_x$, $\hat{L}_y$, and $\hat{L}_z$. If the spin direction is given in terms of the horizontal and azimuthal angles ϕ and θ, the measurement statistics of $\hat{L}(\phi,\theta)$ then read

$$\langle[\hat{L}(\phi,\theta)]^n\rangle = \langle[\sin(\theta)\cos(\phi)\hat{L}_x + \sin(\theta)\sin(\phi)\hat{L}_y + \cos(\theta)\hat{L}_z]^n\rangle. \quad (2)$$

Each nth order expectation value $\langle(\hat{L}_i)^n\rangle$ can therefore be expanded into expectation values of nth order products of the three orthogonal spin components. Specifically, the linear spin expectation values ($n=1$) are defined by the three components of the average spin, $\langle\hat{L}_x\rangle$, $\langle\hat{L}_y\rangle$, and $\langle\hat{L}_z\rangle$, while the quadratic spin expectation values are given by expectation values such as $\langle\hat{L}_x\hat{L}_y+\hat{L}_y\hat{L}_x\rangle$, describing correlations and fluctuations of the spin, and so on. The measurement statistics of any measurement direction can therefore be described by a hierarchy of expectation values ranging from $n=1$ to $n=2l$.

Although there are an infinite number of measurement directions, only a finite number of independent parameters is necessary to describe all nth order expectation values. In general, these independent parameters can be obtained by multiplying out Eq. (2) and identifying the contributions associated with different functions of the angles ϕ and θ. Alternatively, it is possible to identify these contributions with elements of the density matrix. As will be described in more detail below, this corresponds to an expansion of the density matrix into basis operators $\hat{\lambda}_{n,i}$ that can be expressed as nth-order polynomial functions of the spin component operators $\hat{L}_i$. Since the density matrix uniquely determines the spin statistics, the number of independent nth-order expectation values should be equal to the number of independent basis operators $\hat{\lambda}_{n,i}$ in the expansion of the density matrix. The additional density-matrix elements associated with an increase of spin from spin-$(l-1/2)$ to spin-l can then be identified with the additional $(n=2l)$th-order expectation values required to describe the spin statistics given by Eq. (2).

The number of parameters defining the density matrix of any N-level system is N^2-1. A spin-l system is an $N=2l+1$ level system, and the number of parameters defining the density matrix is $4l(l+1)$. A spin-$(l-1/2)$ system is an $N=2l$ level system, and the number of parameters defining the density matrix is $4l^2-1$. The number of additional parameters needed to describe a spin-l system rather than a spin-$(l-1/2)$ system is therefore equal to $4l+1$. Since these additional parameters characterize the $(n=2l)$th order of the measurement statistics given by Eq. (2), the number of independent nth-order expectation values should be equal to $2n+1$. Note that this result is indeed consistent with the number of symmetrically ordered nth-order products of the three spin components required to describe $\langle\hat{L}_n^i\rangle$, once the constant value of the total spin length defined by $\hat{L}_2^x+\hat{L}_2^y+\hat{L}_2^z=l(l+1)$ has been taken into account. Explicit examples for components of the nth-order spin expectation values will be given in Secs. III and V.

TABLE I. Reconstruction of the density matrix from nth-order expectation values of the spin obtained from projective measurements of various spin directions i. The total number of matrix elements necessary for tomography is N^2-1, where $N=2l+1$ is the dimensionality of the corresponding Hilbert space.

	Components of the density matrix					
System size	$\langle\hat{L}_i\rangle$	$\langle\hat{L}_i^2\rangle$	$\langle\hat{L}_i^3\rangle$	$\langle\hat{L}_i^4\rangle$	$\cdots$	Total
Spin-1/2 (two-level)	3				$\cdots$	3
Spin-1 (three -level)	3	5			$\cdots$	8
Spin-3/2 (four-level)	3	5	7		$\cdots$	15
Spin-2 (five-level)	3	5	7	9	$\cdots$	24
$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\cdots$	$\vdots$

Table I illustrates the distribution of density-matrix parameters. Any spin-l system is characterized by $2n+1$ expectation values of nth order, with n running from $n=1$ to $n=2l$. That is, the density matrix can always be represented by three linear spin averages, five second-order spin averages, seven third-order spin averages, and so on [11]. Experimentally, each measurement along a given spin direction determines one nth-order average for each value of n. Since complete quantum tomography requires the determination of $4l+1$ independent contributions to the expectation values of order $n=2l$, it is therefore necessary to measure at least $4l+1$ different spin directions in order to obtain the necessary measurement information [12]. Note that this condition is a result of allowing only measurements of spin components $\hat{L}_i$. If general von Neumann measurements were possible, $2l+2$ measurements would be sufficient for quantum tomography. In the case of spin component measurements, about half of the information obtained is redundant, since it reproduces the results for lower order expectation values that can be obtained from fewer spin directions. For example, quantum tomography of a spin-2 system requires measurements along nine spin directions, providing nine averages to determine the three linear spin expectation values, nine averages to determine the five quadratic spin expectation values, nine averages to determine the seven third-order spin expectation values, and nine averages to determine the nine fourth-order expectation values. Thus it is only the need to determine the complete $(n=2l)$th-order spin statistics that makes it necessary to measure a total of $4l+1$ spin directions $\hat{L}_i$.

Note that the precise choice of measurement directions is not very critical, since the only requirement for obtaining the complete spin statistics is that $2n+1$ of the $4l+1$ averages obtained from the nth-order statistics are linearly independent. In some special cases, the information obtained from $4l+1$ different measurement directions may not be sufficient

to reconstruct the complete density matrix because the choice of measurement directions coincides with a symmetry in the $(n=2l)$th-order expectation values. For example, the second-order information obtained for $\hat{L}_z$ is already obtained from measurements of $\hat{L}_x$ and $\hat{L}_y$, since $\hat{L}_2^x+\hat{L}_2^y+\hat{L}_2^z=l(l+1)$. However, even a small tilt of one of the measurement axes can fix this problem by providing additional information. Therefore, almost any choice of $4l+1$ measurement directions will be sufficient for quantum tomography. To minimize errors, it may be useful to keep the angles between different directions as large as possible [10]. However, the choice of $4l+1$ measurement directions used for a complete reconstruction of the density matrix can generally be quite arbitrary, and each specific tomography protocol merely represents one example out of an infinity of equally valid possibilities.

In order to obtain an explicit description of the density matrix in terms of nth-order spin statistics, it is necessary to identify the contributions of different order n in the density matrix. This can be achieved by expanding the density matrix using an appropriate operator basis. In general, there are infinitely many expansions of the density matrix into orthonormal basis operators. For reasons of mathematical simplicity, the most common choice is that given by the generators of the su(N) algebra [13,14]. However, these generators do not represent the spherical symmetry of the spin-l system. We therefore propose an alternative expansion of the spin-l density matrix $\hat{\rho}_l$ that is based on the different orders n of the spin statistics, given by basis operators $\hat{\lambda}_{n,i}$, where n represents the lowest order of the spin statistics to which $\hat{\lambda}_{n,i}$ contributes, and i is the index of the component within this order, running from 1 to $2n+1$ for each value of n. The conditions for orthogonality and for the normalization of this basis then read

$$\mathrm{Tr}\{\hat{\lambda}_{n,i}\}=0,$$

$$\mathrm{Tr}\{\hat{\lambda}_{n,i}\hat{\lambda}_{m,j}\}=2\delta_{n,m}\ \delta_{i,j}. \tag{3}$$

Using this complete operator basis, it is possible to expand the spin-l density matrix $\hat{\rho}_l$ in terms of expectation values of the spin statistics as

$$\hat{\rho}_l=\frac{1}{2l+1}\hat{1}+\frac{1}{2}\sum_{n=1}^{2l}\left(\sum_{i=1}^{2n+1}\langle\hat{\lambda}_{n,i}\rangle\hat{\lambda}_{n,i}\right). \tag{4}$$

It is thus possible to formulate the density matrix entirely in terms of the measurement statistics of the three-dimensional spin vector, reflecting the analysis of the measurement statistics described by Eq. (2) given in Table I. Specifically, the (N^2-1)-dimensional basis is divided into groups of nth-order products of the spin operators with $2n+1$ operators each, describing the separate nth-order contributions to the spin statistics. In the following, we show how such a basis of three linear and five quadratic spin operators can be defined for the specific case of $l=1$.

III. EXPANSION OF THE SPIN-1 DENSITY MATRIX

The case of $l=1$ provides the most simple example of nonlinear contributions to the spin statistics. Moreover, optical spin-1 systems have already been realized by two photon polarization states [1–4] or by the orbital angular momentum of single photons [5,6]. The following procedure for quantum tomography of a spin-1 system may therefore be particularly useful in the characterization of such experimental results.

For $l=1$, the linear spin components already fulfill the conditions defined by Eqs. (3). The remaining five basis operators can then be constructed using second-order operator products. One such set of quadratic operators that fulfills the relations defined by Eq. (3) [15] is

$$\hat{Q}_{ij}=\hat{L}_i\hat{L}_j+\hat{L}_j\hat{L}_i,$$

$$\hat{S}_{xy}=\hat{L}_x^2-\hat{L}_y^2,$$

$$\hat{G}_z=-\frac{1}{\sqrt{3}}(\hat{L}_x^2+\hat{L}_y^2-2\hat{L}_z^2). \tag{5}$$

In this basis, the generalized eight-dimensional Bloch vector can be separated into a three-dimensional linear part and a five-dimensional quadratic part, given by

$$\hat{\lambda}_{1,1}=\hat{L}_x,\quad \hat{\lambda}_{2,1}=\hat{S}_{xy},$$

$$\hat{\lambda}_{1,2}=\hat{L}_y,\quad \hat{\lambda}_{2,2}=\hat{Q}_{xy},$$

$$\hat{\lambda}_{1,3}=\hat{L}_z,\quad \hat{\lambda}_{2,3}=\hat{Q}_{yz},$$

$$\hat{\lambda}_{2,4}=\hat{Q}_{zx},$$

$$\hat{\lambda}_{2,5}=\hat{G}_z. \tag{6}$$

In the $\hat{L}_z$ basis, the matrix elements of these operators read

$$\hat{\lambda}_{1,1}=\frac{1}{\sqrt{2}}\begin{bmatrix}0&1&0\\1&0&1\\0&1&0\end{bmatrix},\quad \hat{\lambda}_{2,1}=\begin{bmatrix}0&0&1\\0&0&0\\1&0&0\end{bmatrix},$$

$$\hat{\lambda}_{1,2}=\frac{1}{\sqrt{2}}\begin{bmatrix}0&-i&0\\i&0&-i\\0&i&0\end{bmatrix},\quad \hat{\lambda}_{2,2}=\begin{bmatrix}0&0&-i\\0&0&0\\i&0&0\end{bmatrix},$$

$$\hat{\lambda}_{1,3}=\begin{bmatrix}1&0&0\\0&0&0\\0&0&-1\end{bmatrix},\quad \hat{\lambda}_{2,3}=\frac{1}{\sqrt{2}}\begin{bmatrix}0&-i&0\\i&0&i\\0&-i&0\end{bmatrix},$$

$$\hat{\lambda}_{2,4}=\frac{1}{\sqrt{2}}\begin{bmatrix}0&1&0\\1&0&-1\\0&-1&0\end{bmatrix},$$

$$\hat{\lambda}_{2,5} = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 & 0 & 0 \\ 0 & -2 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (7)$$

The expectation values of these eight operators characterize the density matrix in terms of the linear and the quadratic measurement statistics given by Eq. (2). The linear expectation values along any spin direction are given by the three expectation values of $\hat{\lambda}_{1,i}$,

$$\langle \hat{L}(\phi,\theta) \rangle = \sin(\theta)\cos(\phi)\langle \hat{\lambda}_{1,1} \rangle + \sin(\theta)\sin(\phi)\langle \hat{\lambda}_{1,2} \rangle + \cos(\theta)\langle \hat{\lambda}_{1,3} \rangle, \quad (8)$$

and the quadratic expectation values are given by the five expectation values of $\hat{\lambda}_{2,i}$,

$$\begin{aligned} \langle \hat{L}(\phi,\theta)^2 \rangle = {} & \frac{2}{3} + \frac{1}{2}\sin^2(\theta)\cos(2\phi)\langle \hat{\lambda}_{2,1} \rangle \\ & + \sin^2(\theta)\sin(\phi)\cos(\phi)\langle \hat{\lambda}_{2,2} \rangle \\ & + \sin(\theta)\cos(\theta)\sin(\phi)\langle \hat{\lambda}_{2,3} \rangle \\ & + \sin(\theta)\cos(\theta)\cos(\phi)\langle \hat{\lambda}_{2,4} \rangle \\ & + \frac{1}{\sqrt{3}}\left(1 - \frac{3}{2}\sin^2(\theta)\right)\langle \hat{\lambda}_{2,5} \rangle. \end{aligned} \quad (9)$$

Using this relation, it is possible to determine the correct expectation values of all five second-order basis operators $\hat{\lambda}_{2,i}$ from the quadratic expectation values of five independent measurement directions. Together with the linear expectation values, the results of these five measurements then define the complete density matrix,

$$\hat{\rho}_{l=1} = \frac{1}{2}\hat{1} + \underbrace{\frac{1}{2}\sum_{i=1}^{3}\langle \hat{\lambda}_{1,i} \rangle \hat{\lambda}_{1,i}}_{\text{from } \langle \hat{L}(\phi,\theta) \rangle} + \underbrace{\frac{1}{2}\sum_{i=1}^{5}\langle \hat{\lambda}_{2,1} \rangle \hat{\lambda}_{2,i}}_{\text{from } \langle \hat{L}(\phi,\theta)^2 \rangle}. \quad (10)$$

An explicit procedure for quantum tomography can now be formulated by choosing a set of five measurement directions. The expectation values of the basis operators $\hat{\lambda}_{n,i}$ can then be expressed in terms of the measurement probabilities along the five measurement directions.

IV. QUANTUM TOMOGRAPHY OF THE SPIN-1 SYSTEM BASED ON THE MEASUREMENT STATISTICS OF FIVE SPIN DIRECTIONS

For $l=1$, each measurement along a given spin direction $\hat{L}_i$ has three possible outcomes, $m=\pm 1$ and $m=0$. In the experimentally relevant case of two-photon polarization [1–4], these measurement outcomes correspond to the detection of two horizontally polarized photons ($m=+1$), two vertically polarized photons ($m=+1$), and one photon each in horizontal and in vertical polarization ($m=0$), where the component of the Stokes vector is selected by appropriate rotations of the polarization using standard linear optics elements. The measurement statistics given by the probabilities $p_i(m)$ can then be identified with normalized coincident count rates at the detectors.

As discussed above, five measurement settings are necessary to perform quantum tomography. A particularly simple choice of the five measurement directions for the spin-1 system is given by

$$\hat{L}_1 = \hat{L}_x, \quad \hat{L}_2 = \hat{L}_y, \quad \hat{L}_3 = \frac{1}{\sqrt{2}}(\hat{L}_x + \hat{L}_y),$$

$$\hat{L}_4 = \frac{1}{\sqrt{2}}(\hat{L}_y + \hat{L}_z), \quad \hat{L}_5 = \frac{1}{\sqrt{2}}(\hat{L}_z + \hat{L}_x). \quad (11)$$

In the case of two-photon polarization, the first three spin directions can be identified with linear polarization rotated by zero for $\hat{L}_1$, by $\pi/4$ for $\hat{L}_2$, and by $\pi/8$ for $\hat{L}_3$. The remaining two directions then represent elliptical polarizations, with the main axes along $\pm\pi/4$ for $\hat{L}_4$, and the main axes along angles of zero and $\pi/2$ for $\hat{L}_5$. Using this set of measurement settings, it is now possible to explicitly identify the expectation values $\langle \hat{\lambda}_{n,i} \rangle$ that define the density matrix with the corresponding measurement probabilities $p_i(m)$.

In general, the measurement probabilities of a spin-1 system along a given direction i are related to the nth-order expectation values of the corresponding spin component $\hat{L}_i$ by

$$\langle \hat{L}_i \rangle = p_i(+1) - p_i(-1),$$

$$\langle \hat{L}_i^2 \rangle = 1 - p_i(0). \quad (12)$$

The quadratic terms of the spin statistics are thus entirely determined by the measurement probabilities $p_i(0)$ for a measurement value of zero spin along the measurement direction i. The five expectation values $\hat{\lambda}_{2,i}$ defining the quadratic components of the density matrix can therefore be obtained from the five measurement probabilities $p_i(0)$ along the spin directions $i=1$ to $i=5$ defined by Eq. (11). The relations between the measurement statistics and the expectation values of the corresponding basis operators then read

$$\langle \hat{\lambda}_{2,1} \rangle = \langle \hat{S}_{xy} \rangle = -[p_1(0) - p_2(0)],$$

$$\langle \hat{\lambda}_{2,2} \rangle = \langle \hat{Q}_{xy} \rangle = p_1(0) + p_2(0) - 2p_3(0),$$

$$\langle \hat{\lambda}_{2,3} \rangle = \langle \hat{Q}_{yz} \rangle = p_1(0) - 2p_4(0) + 1,$$

$$\langle \hat{\lambda}_{2,4} \rangle = \langle \hat{Q}_{zx} \rangle = p_2(0) - 2p_5(0) + 1,$$

$$\langle \hat{\lambda}_{2,5} \rangle = \langle \hat{G}_z \rangle = \sqrt{3}\left[p_1(0) + p_2(0) - \tfrac{2}{3}\right]. \quad (13)$$

The measurement probabilities for $m=0$ thus determine five of the eight coefficients in the expansion of the density matrix given by Eq. (4). The remaining three coefficients can be

obtained from the linear expectation values given by the differences between $p_i(+1)$ and $p_i(-1)$, e.g.,

$$\langle \hat{\lambda}_{1,1} \rangle = \langle \hat{L}_x \rangle = p_1(+1) - p_1(-1),$$

$$\langle \hat{\lambda}_{1,2} \rangle = \langle \hat{L}_y \rangle = p_2(+1) - p_2(-1),$$

$$\langle \hat{\lambda}_{1,3} \rangle = \langle \hat{L}_z \rangle = -\sqrt{2}\{[p_3(+1) - p_3(-1)] - [p_4(+1) - p_4(-1)] - [p_5(+1) - p_5(-1)]\}. \quad (14)$$

Since the results of five measurement directions are used to determine only three parameters, there are two relations between the measurement results that should be approximately fulfilled if the measurement error is low. These relations can be written as

$$[p_1(+1) - p_1(-1)] \approx \frac{1}{\sqrt{2}}\{[p_3(+1) - p_3(-1)] - [p_4(+1) - p_4(-1)] + [p_5(+1) - p_5(-1)]\},$$

$$[p_2(+1) - p_2(-1)] \approx \frac{1}{\sqrt{2}}\{[p_3(+1) - p_3(-1)] + [p_4(+1) - p_4(-1)] - [p_5(+1) - p_5(-1)]\}. \quad (15)$$

Effectively, the measurement results for the spin directions three to five can be used to predict the results for spin directions one and two. Relations (15) thus illustrate the application of quantum tomography to the prediction of further measurements on the same system. The experimental differences between this prediction and the actual outcome of the measurements may therefore provide a realistic estimate of the errors, limiting the reliability of quantum tomography in practical applications.

Since the eight parameters $\langle \hat{\lambda}_{n,i} \rangle$ completely define the density matrix, they can be used as an alternative representation of the quantum state, just like the three-dimensional Bloch vector for two-level systems. Any other representation of the density matrix can then be obtained from Eq. (4), if the representations of each element of the basis is known. For example, the density-matrix elements in the $\hat{L}_z$ basis given by Eqs. (7) can be used to express any density matrix defined by the expectation values $\langle \hat{\lambda}_{n,i} \rangle$ as

$$\hat{\rho}_{l=1} = \begin{bmatrix} \frac{1}{3} + \frac{1}{2}\langle \hat{\lambda}_{1,3} \rangle + \frac{1}{2\sqrt{3}}\langle \hat{\lambda}_{2,5} \rangle & \frac{1}{2\sqrt{2}}(\langle \hat{\lambda}_{1,1} \rangle - i\langle \hat{\lambda}_{1,2} \rangle - i\langle \hat{\lambda}_{2,3} \rangle + \langle \hat{\lambda}_{2,4} \rangle) & \frac{1}{2}(\langle \hat{\lambda}_{2,1} \rangle - i\langle \hat{\lambda}_{2,2} \rangle) \\ \frac{1}{2\sqrt{2}}(\langle \hat{\lambda}_{1,1} \rangle + i\langle \hat{\lambda}_{1,2} \rangle + i\langle \hat{\lambda}_{2,3} \rangle + \langle \hat{\lambda}_{2,4} \rangle) & \frac{1}{3} - \frac{1}{\sqrt{3}}\langle \hat{\lambda}_{2,5} \rangle & \frac{1}{2\sqrt{2}}(\langle \hat{\lambda}_{1,1} \rangle - i\langle \hat{\lambda}_{1,2} \rangle + i\langle \hat{\lambda}_{2,3} \rangle - \langle \hat{\lambda}_{2,4} \rangle) \\ \frac{1}{2}(\langle \hat{\lambda}_{2,1} \rangle + i\langle \hat{\lambda}_{2,2} \rangle) & \frac{1}{2\sqrt{2}}(\langle \hat{\lambda}_{1,1} \rangle + i\langle \hat{\lambda}_{1,2} \rangle - i\langle \hat{\lambda}_{2,3} \rangle - \langle \hat{\lambda}_{2,4} \rangle) & \frac{1}{3} - \frac{1}{2}\langle \hat{\lambda}_{1,3} \rangle + \frac{1}{2\sqrt{3}}\langle \hat{\lambda}_{2,5} \rangle \end{bmatrix}. \quad (16)$$

The quantum coherences between different eigenstates of $\hat{L}_z$ can thus be expressed in terms of averages involving the other two spin components, $\hat{L}_x$ and $\hat{L}_y$. In fact, this relationship between quantum coherence in the $\hat{L}_z$ basis and the orthogonal spin components $\hat{L}_x$ and $\hat{L}_y$ can be used to systematically construct density-matrix decompositions for higher spins, as we will show in the following.

V. QUANTUM STATISTICS FOR $l > 1$

In order to generalize the construction of a convenient set of basis operators $\hat{\lambda}_{n,i}$ to arbitrarily large spins, it is useful to organize the basis operators according to their density-matrix elements in the $\hat{L}_z$ basis. Such an organization is already indicated by the example for spin-1 given in Eq. (16). In this example, each group of density-matrix elements $|m\rangle\langle m'|$ with the same order of coherence $|m'-m|$ in $\hat{L}_z$ depends on a well-defined subset of the operators $\hat{\lambda}_{n,i}$. This organization of basis operators can be generalized to arbitrary spins l by formulating the operator basis $\hat{\lambda}_{n,i}$ for each spin value in such a way that the matrix elements of each operator are nonzero for only one value of $|m'-m|$. The relationship between the hierarchy of spin expectation values $\langle \hat{\lambda}_{n,i} \rangle$ and the density-matrix elements in the $\hat{L}_z$ basis is then as shown in Table II. Specifically, the $2l+1$ diagonal matrix elements ($|m'-m|=0$) are determined by the set of $2l+1$ operator expectation values $\langle \hat{\lambda}_{n,(2n+1)} \rangle$ that can be obtained from the measurement statistics of $\hat{L}_z$ according to Eq. (1). Likewise, the $4l$ off-diagonal elements with $|m'-m|=1$ can be determined by the set of $4l$ operator expectation values, $\langle \hat{\lambda}_{n,2n} \rangle$ and $\langle \hat{\lambda}_{n,2n-1} \rangle$, and so on. Note that the $4l-2$ off-diagonal elements with $|m'-m|=2$ do not include any first-order spin statistics, since the linear spin components only have matrix elements up to $|m'-m|=1$.

TABLE II. Relation between the density-matrix elements $\langle m'|\hat{\rho}|m\rangle$ in the $\hat{L}_z$ basis and the corresponding nth-order basis operators representing the spin statistics.

	Density-matrix elements							
Spin statistics	$\lvert m'-m\rvert=0$	$\lvert m'-m\rvert=1$	$\lvert m'-m\rvert=2$	$\lvert m'-m\rvert=3$	$\lvert m'-m\rvert=4$	$\lvert m'-m\rvert=5$	…	Total
$\langle\hat{\lambda}_{1,i}\rangle$	1	2					⋯	3
$\langle\hat{\lambda}_{2,i}\rangle$	1	2	2				⋯	5
$\langle\hat{\lambda}_{3,i}\rangle$	1	2	2	2			⋯	7
$\langle\hat{\lambda}_{4,i}\rangle$	1	2	2	2	2		⋯	9
$\langle\hat{\lambda}_{5,i}\rangle$	1	2	2	2	2	2	⋯	11
⋮	⋮	⋮	⋮	⋮	⋮	⋮		⋮
Total	$2l+1$	$4l$	$4l-2$	$4l-4$	$4l-6$	$4l-8$	⋯	$4l(l+1)$

Likewise, the $4l-4$ off-diagonal elements with $|m'-m|=3$ do not include any linear or quadratic spin statistics. In general, an expectation value of at least $|m'-m|$th order is necessary to describe the effect of a corresponding off-diagonal element in the spin statistics.

A convenient way to construct the two operators $\hat{\lambda}_{n,1}$ and $\hat{\lambda}_{n,2}$ with nonzero density-matrix elements of maximal coherence $|m-m'|=n$ is to apply the non-Hermitian spin operator $\hat{L}_x+i\hat{L}_y$. This operator only has nonzero matrix elements with $m'-m=+1$. Specifically,

$$(\hat{L}_x+i\hat{L}_y)|m\rangle=\sqrt{(l-m)(l+m+1)}|m+1\rangle. \tag{17}$$

Consequently, $(\hat{L}_x+i\hat{L}_y)^n$ has only matrix elements with $m'-m=n$. It is therefore possible to generate the nth-order basis operators $\hat{\lambda}_{n,1}$ and $\hat{\lambda}_{n,2}$ with matrix elements of $|m'-m|=n$ from the normalized Hermitian components of $(\hat{L}_x+i\hat{L}_y)^n$,

$$\hat{\lambda}_{n,1}=\frac{(\hat{L}_x+i\hat{L}_y)^n+(\hat{L}_x-i\hat{L}_y)^n}{\sqrt{\mathrm{Tr}\{(\hat{L}_x+i\hat{L}_y)^n(\hat{L}_x-i\hat{L}_y)^n\}}},$$

$$\hat{\lambda}_{n,2}=\frac{-i((\hat{L}_x+i\hat{L}_y)^n-(\hat{L}_x-i\hat{L}_y)^n)}{\sqrt{\mathrm{Tr}\{(\hat{L}_x+i\hat{L}_y)^n(\hat{L}_x-i\hat{L}_y)^n\}}}. \tag{18}$$

Starting from these definitions of basis operators, the complete set of basis operators may be constructed, e.g., by multiplying the operators $\hat{\lambda}_{n,1/2}$ with different powers of $\hat{L}_z$ and/or $\hat{L}_x^2+\hat{L}_y^2$ to obtain higher-order contributions with the same coherence $|m'-m|$ in the density matrix. The precise factors can be determined using the requirements for orthogonality and normalization given by Eq. (3). It is then possible to construct a complete orthonormal operator basis for any spin-l system.

By establishing the relation between spin statistics and coherence in the density matrix, Eq. (18) also illustrates the physical meaning of quantum coherence in $\hat{L}_z$. In particular, it is worth noting that the greater the difference $|m'-m|$ between the $\hat{L}_z$ eigenvalues of the states that are in a coherent superposition, the higher the order of the spin expectation values in $\hat{L}_x$ and $\hat{L}_y$ that is needed to identify this coherence in the measurement statistics. The spin correlation hierarchy presented in Table II may thus provide a key to understanding the non-classical effects associated with quantum superpositions in arbitrarily large physical systems.

VI. NONCLASSICAL CORRELATIONS AND DECOHERENCE

The highest possible value for $|m'-m|$ in the density matrix is obtained for quantum coherence between the extremal $\hat{L}_z$ eigenstates $|m=+l\rangle$ and $|m=-l\rangle$. According to Table II, the two matrix elements describing this coherence correspond to the two $(2l)$th-order spin expectation values. These expectation values can be constructed explicitly using Eq. (18) and include only products of $\hat{L}_x$ and $\hat{L}_y$. In order to characterize a coherent superposition of the $\hat{L}_z$ eigenstates $|m=+l\rangle$ and $|m=-l\rangle$, it is therefore necessary to evaluate the $(2l)$th-order spin statistics in the xy plane. For all orders lower than $2l$, the spin statistics obtained in the measurements of such a coherent superposition are identical to those of an incoherent mixture of $|m=+l\rangle$ and $|m=-l\rangle$.

This observation has significant implications for the identification of strong nonclassical effects in large quantum systems. At sufficiently high values of l, the superposition of $|m=+l\rangle$ and $|m=-l\rangle$ is a (cat-state-like) superposition of two macroscopically distinguishable states. It is therefore interesting to know that the effects of this superposition appear only in the highest-order expectation value of the spin statistics. While the lower-order expectation values are very easy to measure since only very few measurement directions are required and the measurement errors tend to average out, the highest-order expectation values can only be determined from sufficiently precise measurement results of at least $4l+1$ measurement directions. Effectively, the highest-order expectation values represent a measurement resolution at the quantum level, providing the information necessary to resolve the precise eigenvalues of the spin components [16]. This means that the $(2l)$th-order spin statistics is very sensitive to errors of ±1 in the spin measurements. In other words, the smallest measurement errors are sufficient to make the

effects of the cat-state-like superposition between $|m=+l\rangle$ and $|m=-l\rangle$ disappear. We can therefore conclude that the actual nonclassical properties of a superposition of macroscopically distinguishable states can only be observed in the microscopic details of the measurement statistics. It is therefore not surprising that decoherence quickly wipes out such tiny details.

For a more precise evaluation of decoherence and measurement precision, it is useful to consider the case of isotropic decoherence caused by spin diffusion due to random rotations. The time evolution of the density matrix caused by this kind of decoherence can be described by

$$\frac{d}{dt}\hat{\rho}_l = -\Gamma \sum_{i=x,y,z} \left(\frac{1}{2}\hat{L}_i^2\hat{\rho} + \frac{1}{2}\hat{\rho}\hat{L}_i^2 - \hat{L}_i\hat{\rho}\hat{L}_i \right). \tag{19}$$

Using the well-known commutation relations of the spin operators, it is possible to calculate the relaxation dynamics of the nth-order expectation values of the spin. For the non-Hermitian operators $(\hat{L}_x+i\hat{L}_y)^n$ the result reads

$$\frac{d}{dt}\langle(\hat{L}_x+i\hat{L}_y)^n\rangle = -\frac{n(n+1)}{2}\Gamma\langle(\hat{L}_x+i\hat{L}_y)^n\rangle. \tag{20}$$

Since the relaxation of the spin is isotropic, all nth-order contributions to the expansion of the density matrix should relax at the same rate. The effect of isotropic decoherence therefore reduces each nth-order parameter $\langle\hat{\lambda}_{n,i}\rangle$ of the density-matrix expansion (4) by a decoherence factor of $\exp[-\Gamma t n(n+1)/2]$, and the time evolution of the density matrix can be written as

$$\hat{\rho}(t) = \frac{1}{2l+1}\hat{1} + \sum_{n=1}^{2l} \exp\left[-\Gamma t\frac{n(n+1)}{2}\right]\left(\sum_{i=1}^{2n+1}\langle\hat{\lambda}_{n,i}\rangle_{t=0}\hat{\lambda}_{n,i}\right). \tag{21}$$

The expansion of the density matrix $\hat{\rho}_l$ into an operator basis $\hat{\lambda}_{n,i}$ based on the different orders of the spin statistics therefore greatly simplifies the description of any isotropic errors in the preparation and manipulation of spin states.

Since the decoherence effects described by Eq. (21) arise from spin diffusion, it is also possible to identify Γt with an increasing uncertainty in the spin direction, $\Gamma t=\delta\theta^2/2$. The result of Eq. (21) can then be used to estimate the errors caused by a misalignment of the measurement direction. Specifically, an alignment error of $\delta\theta$ will reduce the expectation values observed for the nth-order spin statistics by a factor of $\exp[-\delta\theta^2 n(n+1)/4]$. To obtain at least $\exp[-0.25]=78\%$ of the original expectation value at orders $n\gg 1$ of the spin statistics, the errors of the spin alignment have to be smaller than $\delta\theta=1/n$. The precision in the alignment of the spin direction necessary to obtain the nth-order statistics is thus proportional to $1/n$, and the requirement for observing evidence of catlike superpositions in spin-l systems is an angular resolution of $\delta\theta<1/(2l)$.

VII. ENTANGLEMENT STATISTICS AND GENERAL SPIN NETWORKS

The formalism developed above can also be applied to entangled spin-l systems. In this case, the density matrix of the total system is obtained by evaluating the correlations between measurements of the local spin components. Specifically, the joint quantum state of a spin-l_A system A and a spin-l_B system B can be determined by simultaneously measuring spin components $\hat{L}_i(A)$ in A and spin components $\hat{L}_i(B)$ in B, obtaining the joint probabilities $p_{ij}(m_A,m_B)$ of each measurement outcome. The correlated spin statistics can then be expressed in terms of the expectation values,

$$\langle[\hat{L}_i(A)]^{n_A}\otimes[\hat{L}_i(A)]^{n_B}\rangle = \sum_{m_A,m_B} m_A^{n_A}m_B^{n_B}p_{ij}(m_A,m_B). \tag{22}$$

It is then possible to analyze the spin statistics according to the local order n_A and n_B, where the total number of independent components required to characterize each order is given by the product $(2n_A+1)(2n_B+1)$. Note that in this case, $n_A=0$ and $n_B=0$ have to be included in order to describe the local spin statistics of each system. Consequently, the lowest order expectation values are given by $(n_A=1,n_B=0)$ and $(n_A=0,n_B=1)$, with three independent components each. The second-order expectation values $(n_A+n_B=2)$ are given by five components for $(n_A=2,n_B=0)$, nine components for $(n_A=1,n_B=1)$, and five components for $(n_A=0,n_B=2)$. The highest order contribution to the correlated spin statistics is then given by $(n_A=2l_A,n_B=2l_B)$, with a total of $(4l_A+1)(4l_B+1)$ independent components. The number of measurement settings required to perform complete quantum tomography for entangled spin systems is therefore equal to $(4l_A+1)(4l_B+1)$. In the experimentally realized case of $l_A=l_B=1$ [3–6], this would require 25 different measurement settings with nine possible outcomes each, for a total of 225 measurement probabilities.

An explicit description of the density matrix in terms of the correlated (n_A,n_B)th-order spin statistics can be obtained using products of the basis operators for each individual system. The expansion of the density matrix then reads

$$\begin{aligned}\hat{\rho}_{AB} = {} & \frac{1}{(2l_A+1)(2l_B+1)}\hat{1}\otimes\hat{1} + \frac{1}{2(2l_A+1)}\sum_{n_A}\left(\sum_i\langle\hat{\lambda}_{n_A,i}\right. \\ & \left.\otimes\hat{1}\rangle\hat{\lambda}_{n_A,i}\otimes\hat{1}\right) + \frac{1}{2(2l_B+1)}\sum_{n_B}\left(\sum_i\langle\hat{1}\otimes\hat{\lambda}_{n_B,i}\rangle\hat{1}\right. \\ & \left.\otimes\hat{\lambda}_{n_B,i}\right) + \frac{1}{4}\sum_{n_A,n_B}\left(\sum_{i,j}\langle\hat{\lambda}_{n_A,i}\otimes\hat{\lambda}_{n_B,j}\rangle\hat{\lambda}_{n_A,i}\otimes\hat{\lambda}_{n_B,j}\right).\end{aligned} \tag{23}$$

The expectation values defining the density matrix can now be expressed in terms of the joint measurement probabilities $p_{ij}(m_A,m_B)$ by writing the (n_A,n_B)th-order expectation values of the correlated spins in Eq. (22) as a function of the expectation values in Eq. (23). It is then possible to fully characterize any $N\times M$ entanglement in terms of the correlated spin statistics.

The extension of this formalism to multipartite spin networks is also straightforward, since the density matrix can be expanded into products of the local basis operators for any number of systems. The expectation values of these products can then be determined from the correlated measurement statistics of spin measurements performed simultaneously on all systems.

VIII. CONCLUSIONS

In conclusion, we have shown how the density matrix of spin-l systems can be reconstructed from the measurement statistics of projective spin measurements along a set of at least $4l+1$ different spin directions. The components of the density matrix can then be identified with different contributions to the statistics of the three-dimensional spin vector. It is therefore possible to interpret the discrete quantum statistics of arbitrarily large spin systems within the same three-dimensional space defined by the Bloch vector of a two-level system.

The explicit procedure for the quantum tomography of spin-1 systems provides an example of the general method that can be applied directly to experimentally generated two-photon polarization states such as the ones reported in Refs. [1–4]. It may thus serve as the foundation of a more detailed characterization of decoherence and noise effects in these newly available entanglement sources.

Besides its practical usefulness for the experimental characterization of general spin-l systems, the expansion of the density matrix into elements of the spin statistics also provides a more intuitive understanding of quantum statistics in large systems. The analysis presented above may therefore also help to clarify the conditions for the emergence of quantum effects in physical systems of arbitrary size.

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[1] T. Tsegaye, J. Soderholm, M. Atature, A. Trifonov, G. Bjork, A. V. Sergienko, B. E. A. Saleh, and M. C. Teich, Phys. Rev. Lett. **85**, 5013 (2000).
[2] A. V. Burlakov, M. V. Chekhova, O. A. Karabutova, and S. P. Kulik, Phys. Rev. A **64**, 041803(R) (2001).
[3] A. Lamas-Linares, J. C. Howell, and D. Bouwmeester, Nature (London) **412**, 887 (2001).
[4] J. C. Howell, A. Lamas-Linares, and D. Bouwmeester, Phys. Rev. Lett. **88**, 030401 (2002).
[5] A. Mair, A. Vaziri, G. Weihs, and A. Zeilinger, Nature (London) **412**, 313 (2001).
[6] A. Vaziri, G. Weihs, and A. Zeilinger, Phys. Rev. Lett. **89**, 240401 (2002).
[7] A. G. White, D. F. V. James, P. H. Eberhard, and P. G. Kwiat, Phys. Rev. Lett. **83**, 3103 (1999).
[8] D. F. V. James, P. G. Kwiat, W. J. Munro, and A. G. White, Phys. Rev. A **66**, 052312 (2001).
[9] R. T. Thew, K. Nemoto, A. G. White, and W. J. Munro, Phys. Rev. A **66**, 012303 (2002).
[10] G. Klose, G. Smith, and P. S. Jessen, Phys. Rev. Lett. **86**, 4721 (2001).
[11] It may be worth noting that, in terms of group theory, the $2n+1$ expectation values of nth-order forms an irreducible representation of the spherical symmetry defined by the arbitrary direction of the spin component $\hat{L}_i$ in Eq. (2).
[12] It has recently come to our attention that a particular choice of $4l+1$ measurement directions was proposed as early as 1968 by R. G. Newton and B.-L. Young, Ann. Phys. (N.Y.) **49**, 393 (1968). The results in this paper show that $4l+1$ measurement directions are also sufficient for quantum tomography. However, Newton and Young suggested that a better choice of directions could reduce the number of measurement directions needed. Our results show that this is in fact impossible.
[13] F. T. Hioe and J. H. Eberly, Phys. Rev. Lett. **47**, 838 (1981).
[14] G. Mahler and V. A. Weberruß, *Quantum Networks* (Springer, Berlin, 1998), p. 44.
[15] H. F. Hofmann, Phys. Rev. A **68**, 034307 (2003).
[16] In fact, the truncation of the measurement statistics at $\langle(\hat{L}_i)^{2l}\rangle$ itself is a consequence of the fact that the discreteness of the quantized eigenvalues makes a higher resolution unnecessary.