

Pure and mixed state calculations of the laser-induced ionization of uranium

I J Ford

Theoretical Studies Department, AEA Industrial Technology, B424.4, Harwell Laboratory, Didcot, Oxon OX11 0RA, UK

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Abstract. In calculations of the laser-induced ionization of uranium atoms it is necessary to average over all phases in the initial multicomponent wavefunction, the components corresponding to the initially occupied hyperfine states. In practice, however, it has been found that the results of such a mixed state calculation differ only slightly from those obtained from a pure state calculation, which uses a single initial state with a random choice of phases. The statistical properties of multidimensional unit vectors are used to explain this observation, suggesting that the system is sufficiently complicated that the properties of the eigenvectors of the Hamiltonian are well represented by an ensemble over all possible orientations. This supports the use of pure state calculations in laser isotope separation studies, which offer a large reduction in the necessary computational effort.

1. Introduction

The separation of the isotopes of uranium has major commercial importance in the nuclear fuel industry. As is well known, naturally occurring uranium contains only 0.72% of the fissile ^{235}U isotope, the remainder being mostly the non-fissile and neutron-absorbing isotope ^{238}U . The enrichment of the ^{235}U fraction is necessary for most current nuclear reactor designs, and represents a major component of the effort and cost of the production of nuclear fuel.

Most enrichment is carried out with gas centrifuges or diffusion plants, exploiting the small difference in the masses of the two isotopes. Since this leads to very small differences in physical properties such as the diffusion rate, the process necessarily involves cascades of large numbers of devices in order to produce a significant enrichment. The capital cost of the plant involved is large, and more importantly, the energy needed to run the machines and produce a given enrichment is high. For this reason, alternative methods for separating the elements of uranium are being studied, notably the use of lasers to induce preferential ionization of ^{235}U (see Greenland 1991 for a review).

Laser isotope separation relies on differences between the electronic energy levels of ^{235}U and ^{238}U . These differences are mainly due to the slightly larger nuclear radius for ^{238}U , which means that the electrons see less of the nuclear potential and are consequently less strongly bound than in ^{235}U . Both the binding energies and the energies of atomic transitions differ. The situation is complicated by the effect of nuclear spin, which leads to different hyperfine structures within the spectra of the two isotopes. The nuclear spin of ^{235}U is $\frac{7}{2}$, so that each transition in the ^{235}U spectrum is split into an octet. The nuclear spin of ^{238}U is zero, however, so the transitions are singlets. In spite of the spreading of the ^{235}U spectrum due to the hyperfine structure, though, the nuclear radius effect is strong enough

to separate the transitions in ^{235}U from those in ^{238}U . This is exploited using a laser with a bandwidth smaller than the frequency separation of the spectra, tuned to excite transitions in ^{235}U but not in ^{238}U . A very effective separation of the isotopes is then possible by ionizing and collecting the ^{235}U ions electromagnetically.

With lasers currently available, it is not possible to ionize the ^{235}U isotope in a single step, but instead a multiphoton process is envisaged, involving a ladder of atomic transitions. These can be arranged linearly, or in more complicated arrangements, depending on the situation. Possible ladders are illustrated in figure 1. The laser frequencies chosen, and therefore the route taken by the ladder through the very complicated uranium spectrum (Ahmad and Pandey 1980), is a matter of great importance. This is because the ionization depends on the various transition strengths involved. Also, loss of population from the ladder by spontaneous decay to other levels should be minimized. In order to optimize the choice of ladder, a theoretical description of the ionization process can be very useful. The problem can be described by a Schrödinger equation for the evolution of a wavefunction in a space spanned by the N coupled atomic states. The calculation requires a set of atomic parameters, such as decay rates, coupling strengths to the laser field, and hyperfine splittings, together with a set of laser parameters, including frequencies, intensities, pulse shapes, phases and delays. The calculation of ionization yield can then be performed by simply evolving the atomic system from an initial state through the excitation process.

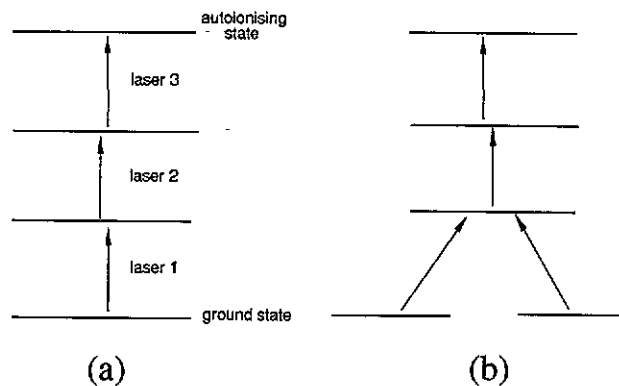


Figure 1. Ladder schemes for the excitation of uranium. A multiphoton process is envisaged, involving the sequential excitation of an electron from one (a) or more (b) initial states through several levels to an autoionizing top level.

Although conceptually straightforward, the calculation is extremely complex, since the interactions are time dependent and a large number of effects need to be taken into account, including Doppler broadening and Zeeman splitting. A further complication is the number of states involved: the ground state of ^{235}U has an orbital angular momentum J equal to 6, which together with the nuclear spin I of $\frac{7}{2}$ leads to a multiplicity of the ground state atomic level of $(2J + 1)(2I + 1) = 104$. For a typical ladder consisting of three laser frequencies, coupling four atomic levels, this implies the coupling of over 400 states, though selection rules reduce this estimate to some extent. Numerical evolution of the quantum mechanical state according to the details of the laser interactions then becomes very time consuming and costly. It is therefore sensible to look for ways in which the complexity of the calculation can be reduced, while still retaining a sufficiently realistic description of the ionization process. A scheme for doing so is examined in this paper.

An exact quantum mechanical calculation of the excitation process involves taking an average over all possible initial states, which is known as a mixed state calculation. However, we have noticed that in our uranium ionization case a pure state calculation, which is an evolution from a particular initial state, gives results which differ only slightly from exact computations. This can greatly reduce the scale of the problem. However, the saving can only be exploited with confidence if the reason for the similarity is understood. This paper attempts to define the circumstances which lead to similarities in the pure and mixed state calculations.

An analysis of pure and mixed state calculations for the excitation of uranium is made in the next section, using statistical properties of eigenvectors to estimate the different ionization yields. Section 3 gives an illustration of the possible computational savings and a summary of the work and conclusions are given in section 4. An appendix describes the statistics of multidimensional unit vectors.

2. Pure and mixed state calculations

Quantum mechanical calculations starting from either a pure or a mixed initial state are most easily compared for a simplified case. We consider a situation where the laser fields are switched on at time $t = 0$ and operate at constant frequency and amplitude for an interval T . We neglect transitions to states outside the ladder of atomic levels considered. The corresponding Hamiltonian in a suitable set of basis states and using the rotating wave approximation, is real and time independent, and describes all atomic and atom-laser interactions. The effects of going beyond these assumptions are examined later.

Consider a set of N states coupled by a Hamiltonian H . The system is described by a normalized N -component wavefunction ψ_l ($l = 1, N$). The evolution from an initial state denoted ψ^0 is written

$$\psi_j = U_{jk} \psi_k^0 \quad (1)$$

where U is the evolution operator, given by

$$U = \exp(-iHt). \quad (2)$$

If H is time independent, then the matrix elements of U are given by

$$U_{jk} = \sum_i u_j^i \exp(-i\lambda_i t) u_k^i \quad (3)$$

where λ_i is the i th eigenvalue and u_j^i is the j th component of the i th eigenvector of H . The N eigenvectors form an orthogonal set of real unit vectors in N -dimensional space. The population at time t in the state j is then given by

$$|\psi_j(t)|^2 = \sum_{k,m} \sum_{i,l} u_j^i u_k^i u_j^l u_m^l \exp(-i(\lambda_i - \lambda_l)t) \psi_k^0 \psi_m^{0*}. \quad (4)$$

We shall be interested in the mean population found in a specified subset of states $\{\psi_j\}$ over the interaction period T . These represent the states from which ionization would occur, although no losses are yet allowed for in the Hamiltonian. In the limit of very small decay

rates, the time-integrated population in the j states divided by the lifetime τ would yield the fractional ionization. The mean population in the j states is

$$\frac{1}{T} \sum_j \int_0^T |\psi_j|^2 dt = \sum_j \sum_{k,m} \sum_{i,l} u_j^i u_k^i u_j^l u_m^l \left(\frac{(1 - \exp(-i(\lambda_i - \lambda_l)T))}{i(\lambda_i - \lambda_l)T} \right) \psi_k^0 \psi_m^{0*} \quad (5)$$

$$= \sum_j \sum_{k,m} \sum_i u_j^i u_k^i u_j^i u_m^i \psi_k^0 \psi_m^{0*} + \sum_j \sum_{k,m} \sum_{i \neq l} u_j^i u_k^i u_j^l u_m^l \times \left[\frac{1 - \cos(\lambda_i - \lambda_l)T}{(\lambda_i - \lambda_l)T} \text{Im}(\psi_k^0 \psi_m^{0*}) + \frac{\sin(\lambda_i - \lambda_l)T}{(\lambda_i - \lambda_l)T} \text{Re}(\psi_k^0 \psi_m^{0*}) \right]. \quad (6)$$

The eigenvalues λ_i represent oscillation frequencies of population transfer amongst the states. It is assumed that no two states are exactly degenerate. If many such oscillations take place during the excitation interval, i.e. $(\lambda_i - \lambda_l)T \gg 1$, then the second sum in equation (6) can be neglected. (In practice, T is of the order of tens of ns, and the difference in frequencies a few hundreds of MHz, so that this approximation is reasonable.) The resulting averaged excited population is time independent, within the approximation taken, which suggests that an approach using time-independent perturbation theory might have yielded the same result.

Consider a calculation starting from a situation where population is divided between a subset $\{\psi_k\}$ of the states. These would correspond to the initially occupied states of the atom. We shall insist that the j states and the k states are separate subsets of the N coupled states. Let the initial wavefunction be

$$\psi_k^0 = a_k \exp(i\alpha_k) \quad (7)$$

where the $\{a_k\}$ are a suitably normalized set of amplitudes and the $\{\alpha_k\}$ are a set of phases. From equation (6), we have to a good approximation,

$$\frac{1}{T} \sum_j \int_0^T |\psi_j|^2 dt = I_p = \sum_j \sum_{k,m} \sum_i a_k a_m u_j^i u_k^i u_j^i u_m^i \exp i(\alpha_k - \alpha_m) \quad (8)$$

which defines the pure state mean excited population I_p .

The specified initial population in each of the k states defines the amplitudes a_k but the phases are undetermined. The mixed state calculation performs an average over all possible phase relationships in the initial state. Thus from equation (8) the mixed state mean excited population I_m is given by

$$I_m = \sum_j \sum_k \sum_i a_k^2 u_j^i u_k^i u_j^i u_k^i. \quad (9)$$

so that, defining an error E ,

$$I_p = I_m + E = I_m + \sum_j \sum_{k \neq m} \sum_i a_k a_m u_j^i u_k^i u_j^i u_m^i \cos(\alpha_k - \alpha_m). \quad (10)$$

These results can be applied to the case in question. Exact calculations of the ionization of a uranium atom require an average over the initial phases in the wavefunction, which is equivalent to performing separate evolutions of the system starting with all the initial

population in just one of the k states. Thus the effort required to perform a mixed state calculation is nominally N_k times that of a pure state calculation, where N_k is the number of initially populated k states.

Numerical codes have been written which calculate the ionization of uranium (Lauder and Greenland 1984) and which can use either pure or mixed state initial conditions. The pure initial state can have either a particular choice of phases, or a random set. In practice, it is found that the differences in computed ionization yields differ only slightly in all cases examined, while there is a large reduction in effort for the pure state calculation compared with the mixed state. The insensitivity may be partly ascribed to saturation of the ionization yield: for the laser parameters of interest commercially, each possible excitation route provides nearly complete ionization, which is therefore insensitive to the choice of initial conditions. However, even for low ionization rates, well away from optimum conditions, the differences are small. This suggests that the similarity in the results of the pure and mixed state calculations is in some way intrinsic to the system, and that a great saving can be made in calculating ionization yields, at the cost of introducing a small error relative to the exact calculation. The saving can only be exploited, however, if some estimate of the likely error can be made.

The result given in equation (10) allows one to discuss the difference, E , between the pure and mixed state calculations in analytical terms. The expressions depend on the orthonormal set of real eigenvectors $\{u^i\}$ of H spanning the N -dimensional space. The orientation of the eigenvectors in the space depends on the details of the Hamiltonian. Recall that the system described so far is time independent, so the eigenvectors are fixed in this space. When time dependence is introduced, they will move as the Hamiltonian changes. The idea that we shall pursue in the remainder of this section is that a real system approximates to the average behaviour of an ensemble of randomly oriented eigenvectors in the N -dimensional space. It is likely that this situation is favoured by an increasing complexity of the system and an increasing dimension N . This hypothesis will lead to numerical estimates of E which can be compared against actual pure/mixed state calculational differences.

Consider the mean properties of a set of random unit vectors in N dimensions, neglecting the orthogonality requirement. We are interested in the pure and mixed state mean excited populations $\langle I_p \rangle$ and $\langle I_m \rangle$ respectively, and the statistics of the error E , obtained by averaging within such an ensemble. We have

$$\langle I_m \rangle = \sum_j \sum_k \sum_i a_k^2 \langle u_j^{i2} u_k^{i2} \rangle_N \quad (11)$$

where the subscript N denotes the dimensionality of the eigenvectors. In the appendix it is shown that for $j \neq k$, the moment in equation (11) is given by $1/(N(N+2))$ so that

$$\langle I_m \rangle = \sum_j \sum_k a_k^2 \frac{1}{(N+2)}. \quad (12)$$

Similarly, $\langle E \rangle = 0$ and

$$\begin{aligned} \langle E^2 \rangle &= \sum_j \sum_{k \neq m} \sum_i a_k^2 a_m^2 \langle u_j^{i4} u_k^{i2} u_m^{i2} \rangle_N \cos^2(\alpha_k - \alpha_m) \\ &= \sum_j \sum_{k \neq m} a_k^2 a_m^2 \frac{3}{(N+2)(N+4)(N+6)} \cos^2(\alpha_k - \alpha_m) \end{aligned} \quad (13)$$

using a result given in the appendix.

Equations (12) and (13) are the basic relations that we use to estimate the error introduced by a pure state calculation, but let us first write them in a form more suited to the calculations with uranium. At present, ionization calculations are required only for situations where m_F , the z component of the total angular momentum $F = I + J$, is a constant of the motion. This leads to a separation of the excitation problem into a number of parallel ladders, labelled by p , involving a set of coupled hyperfine states with the same m_F . The initial occupation amplitudes $\{a_k\}$ may be written as $(v_k/d_k)^{1/2}$ where v_k is the fraction of the population initially in the atomic level to which the state k belongs, and d_k is the $(2J + 1)(2I + 1)$ multiplicity of that level. Then

$$\langle I_m \rangle = \sum_p \sum_{j_p} \sum_{k_p} \frac{v_{k_p}}{d_{k_p}} \frac{1}{(N_p + 2)} \quad (14)$$

and

$$\langle E^2 \rangle = \sum_p \sum_{j_p} \sum_{k_p \neq m_p} \frac{v_{k_p}}{d_{k_p}} \frac{v_{m_p}}{d_{m_p}} \frac{3}{(N_p + 2)(N_p + 4)(N_p + 6)} \cos^2(\alpha_{k_p} - \alpha_{m_p}), \quad (15)$$

where k_p etc label the states within each excitation ladder. Equation (14) may be written

$$\langle I_m \rangle = \sum_p \sum_l \frac{v_l}{d_l} N_{fp} N_{ilp} \frac{1}{(N_p + 2)} \quad (16)$$

where now the subscripts to v and d refer to the atomic level l . N_{fp} is the number of (final) j states in the excitation ladder labelled p and N_{ilp} is the number of (initial) k states in atomic level l and ladder p . Equation (15) can be similarly rewritten, at the same time performing a further averaging over the phases $\{\alpha_k\}$:

$$\begin{aligned} \overline{\langle E^2 \rangle} &= \sum_p N_{fp} \left(\sum_l \frac{v_l^2}{d_l^2} N_{ilp} (N_{ilp} - 1) + \sum_{n \neq l} \frac{v_l}{d_l} \frac{v_n}{d_n} N_{ilp} N_{inp} \right) \\ &\times \frac{3}{2(N_p + 2)(N_p + 4)(N_p + 6)} \end{aligned} \quad (17)$$

where n and l label atomic levels.

These expressions can be simplified in the case of one initially occupied atomic level, for which $v_1=1$ and $v_l=0$ for $l \neq 1$. The ratio of expected error to the correct result is

$$\frac{(\overline{\langle E^2 \rangle})^{1/2}}{\langle I_m \rangle} = \left(\sum_p \frac{3N_{ip}(N_{ip} - 1)N_{fp}}{2(N_p + 2)(N_p + 4)(N_p + 6)} \right)^{1/2} \left(\sum_p \frac{N_{ip}N_{fp}}{(N_p + 2)} \right)^{-1} \quad (18)$$

where we have dropped the l subscript on the N_{ip} . This is the expression we shall use to estimate the relative error introduced by the pure state calculation.

Table I gives a set of multiplicities N_{ip} and N_{fp} for 'initial' and 'final' states respectively in a set of ladders labelled by different m_F for a system of four $J = 6$ atomic levels with $I = \frac{7}{2}$ and linearly polarized lasers ($\Delta m_F = 0$). The states are coupled as in figure 1(a). The total dimensionality N_p of each coupled sub-system is also shown, together with the fractional error for each sub-system and for the whole problem, calculated using equation (18). The expected difference between the pure and mixed state calculations is of the order of 2% for this typical system, on the basis of the statistical properties of unit N -dimensional vectors. In the next section this estimate is compared with some example numerical calculations, in order to test the hypothesis that the average properties of the eigenvectors represent the behaviour of real systems.

Table 1. Properties of ladders of constant m_F for a four-level $J = 6$, $I = \frac{7}{2}$ system, together with estimates of differences between pure and mixed state calculations, given by equation (18).

| m_F | N_{iP} | N_{fP} | N_P | Error (%) |
|-------|----------|----------|-------|-----------|
| -19/2 | 1 | 1 | 4 | 0 |
| -17/2 | 2 | 2 | 8 | 14.9 |
| -15/2 | 3 | 3 | 12 | 12.7 |
| -13/2 | 4 | 4 | 16 | 10.7 |
| -11/2 | 5 | 5 | 20 | 9.2 |
| -9/2 | 6 | 6 | 24 | 8.0 |
| -7/2 | 7 | 7 | 28 | 7.1 |
| -5/2 | 8 | 8 | 32 | 6.4 |
| -3/2 | 8 | 8 | 32 | 6.4 |
| -1/2 | 8 | 8 | 32 | 6.4 |
| 1/2 | 8 | 8 | 32 | 6.4 |
| 3/2 | 8 | 8 | 32 | 6.4 |
| 5/2 | 8 | 8 | 32 | 6.4 |
| 7/2 | 7 | 7 | 28 | 7.1 |
| 9/2 | 6 | 6 | 24 | 8.0 |
| 11/2 | 5 | 5 | 20 | 9.2 |
| 13/2 | 4 | 4 | 16 | 10.7 |
| 15/2 | 3 | 3 | 12 | 12.7 |
| 17/2 | 2 | 2 | 8 | 14.9 |
| 19/2 | 1 | 1 | 4 | 0 |
| Total | | | | 1.87 |

Table 2. Pure and mixed state calculations of ionization yield for the system considered in table 1. The pure state results are obtained using random phases.

| | Yield |
|--------------------|--------|
| Mixed state | 0.3846 |
| Pure state | 0.3811 |
| | 0.4028 |
| | 0.3849 |
| | 0.3872 |
| | 0.3831 |
| | 0.3909 |
| | 0.3702 |
| | 0.3872 |
| Mean | 0.3859 |
| Standard deviation | 0.0092 |

3. Numerical examples

We consider a four-level ladder similar to scheme (a) shown in figure 1 with $J = 6$ for each level, and linearly polarized lasers, which separate the problem into 20 separately coupled sub-systems with constant m_F , as described in table 1. Decay from all levels except the lowest, and time dependent interactions are included. The couplings, hyperfine splittings, laser detunings, magnetic field and pulse shapes, etc, have been chosen to produce an ionization yield from the top level in the region of 40%, and do not need to be specified here.

A mixed state calculation gives the exact ionization yield for the system. On the other hand, a number of pure state calculations, using phases $\{\alpha_k\}$ chosen randomly from

a uniform distribution in $0 \leq \alpha_k \leq 2\pi$, gives a scatter of results. From these, a mean and standard deviation can be calculated. Results from this analysis are given in table 2. The difference between the mixed state yield and a pure state yield based on a randomly chosen set of phases is clearly small and of about the same size as the error derived in the previous section. In this particular case the differences are about 2.4%. The mixed state calculation took 102 CPU s on one processor of a Cray-2 supercomputer, however, while each pure state calculation took 42 CPU s. This is a gain of about 2.4 in computation time which represents a valuable reduction in effort. On scalar machines the acceleration has been found to be even greater, but the acceleration is not as large as the nominal factor of 8 (the number of initially populated states) expected from earlier considerations. This is because the computations involve overheads which are independent of whether a pure or a mixed initial state is used, so that the gain will necessarily fall short of this maximum.

4. Discussion and conclusions

The task of calculating the ionization yield due to the multiphoton excitation of a complicated atom such as uranium is made much easier by using a pure state rather than a mixed state for the initial condition. The mixed state calculation is more precise since it performs an average over all possible phase differences between the components of the initial wavefunction. These components correspond to various hyperfine states in the initially populated atomic levels. A pure state calculation is based on a particular set of phases, and involves less computational effort since an averaging procedure is not necessary. In practice, we have found that the ionization yields resulting from pure and mixed state calculations are quite similar, becoming more so with more complicated systems and larger fractional ionizations. We have suggested that this similarity may be understood by considering the average properties of the eigenvectors of the Hamiltonian. This is equivalent to calculating the excitation of a system using all possible orientations of the eigenvectors, and taking averages.

Using the statistical properties of multidimensional unit vectors, the expected error introduced by the pure state calculation has been estimated. This estimate gives the correct magnitudes of deviations between the two cases found in an example calculation. Although the ionization yields themselves are not accounted for numerically by the calculations based on the eigenvector ensemble, the relative magnitudes of the pure and mixed calculations are, which makes the approach useful. Pure state calculations can now be accompanied by an estimate of the probable deviation from the exact mixed state result.

A number of details of the analysis need to be considered. In the development in section 2, The Hamiltonian was taken to be time independent and decay to states outside the ladder was neglected. These assumptions need not invalidate the development if population changes between levels occur on a faster timescale than either of these effects, which will almost always apply. Furthermore, population oscillations typically take place over periods much shorter than the pulse length, so the neglect of the $i \neq l$ terms in equation (6) is justified. The assumption that there is no degeneracy within the set of eigenvalues will probably hold when considering a complicated system with many interactions.

We conclude that the laser-induced ionization yield of ^{235}U is insensitive to the choice of phases in the initial state, for conditions of interest in laser isotope separation, and that this can be exploited to reduce the effort necessary in performing calculations. The reason for the insensitivity is that the system is sufficiently complicated that the properties of the eigenvectors of the Hamiltonian can be represented by those of an ensemble. It would be of

interest to study other systems in order to determine what particular features are important in bringing about this insensitivity.

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Appendix

We here describe the statistics of a unit vector in N dimensions. The probability distribution of the components $\{x_i\}$ of the vector is

$$p(\{x_i\}) = \frac{\delta\left(\sum_{i=1}^N x_i^2 - 1\right)}{\int \delta\left(\sum_{i=1}^N x_i^2 - 1\right) \prod_{i=1}^N dx_i}.$$

The delta function ensures that the vector has unit length and the probability distribution is correctly normalized. The second moment of the i th component of the vector is

$$\begin{aligned} \langle x_i^2 \rangle_N &= \frac{\int x_i^2 \delta\left(\sum_{i=1}^N x_i^2 - 1\right) \prod_{i=1}^N dx_i}{\int \delta\left(\sum_{i=1}^N x_i^2 - 1\right) \prod_{i=1}^N dx_i} \\ &= \frac{1}{N} \frac{\int \sum_{i=1}^N x_i^2 \delta\left(\sum_{i=1}^N x_i^2 - 1\right) \prod_{i=1}^N dx_i}{\int \delta\left(\sum_{i=1}^N x_i^2 - 1\right) \prod_{i=1}^N dx_i} \\ &= \frac{1}{N} \end{aligned}$$

since for any y , $y\delta(y-1)$ can be replaced by $\delta(y-1)$ within an integral. The subscript N on the expectation value denotes the dimensionality of the vector.

Also, for $N \geq 4$,

$$\begin{aligned} \langle x_j^2 \rangle_N &= \frac{\int \prod_{i=1}^{N-3} dx_i r^2 dr d\Omega x_j^2 \delta\left(\sum_{i=1}^{N-3} x_i^2 + r^2 - 1\right)}{\int \prod_{i=1}^{N-3} dx_i r^2 dr d\Omega \delta\left(\sum_{i=1}^{N-3} x_i^2 + r^2 - 1\right)} \\ &= \frac{\langle x_i^2 x_j^2 \rangle_{N-2}}{\langle x_i^2 \rangle_{N-2}} \quad i \neq j \end{aligned}$$

where $r^2 dr d\Omega$ is the volume element involved in the integration over the three components (x_{N-2}, x_{N-1}, x_N) , with $j \leq N-3$. Therefore

$$\langle x_i^2 x_j^2 \rangle_N = \frac{1}{N(N+2)} \quad i \neq j, N \geq 2.$$

Now, the fourth moment of x_i can be written

$$\begin{aligned}\langle x_i^4 \rangle_N &= \frac{1}{N} \left\langle \left(\sum_{i=1}^N x_i^4 \right) \right\rangle_N \\ &= \frac{1}{N} \left(\left\langle \left(\sum_{i=1}^N x_i^2 \right)^2 \right\rangle_N - \sum_{i \neq j} \langle x_i^2 x_j^2 \rangle_N \right) \\ &= \frac{1}{N} \left(1 - \frac{N(N-1)}{N(N+2)} \right) \\ &= \frac{3}{N(N+2)}\end{aligned}$$

using previous results. Higher moments can be calculated by similar means. In particular, we write, for $N \geq 4$

$$\begin{aligned}\langle x_j^4 \rangle_N &= \frac{\int \prod_{i=1}^{N-3} dx_i r^2 dr d\Omega x_j^4 \delta \left(\sum_{i=1}^{N-3} x_i^2 + r^2 - 1 \right)}{\int \prod_{i=1}^{N-3} dx_i r^2 dr d\Omega \delta \left(\sum_{i=1}^{N-3} x_i^2 + r^2 - 1 \right)} \\ &= \frac{\langle x_i^4 x_j^2 \rangle_{N-2}}{\langle x_i^2 \rangle_{N-2}} \quad i \neq j.\end{aligned}$$

So

$$\langle x_i^4 x_j^2 \rangle_N = \frac{3}{N(N+2)(N+4)}.$$

The previous step can be repeated:

$$\begin{aligned}\langle x_i^4 x_j^2 \rangle_N &= \frac{\int \prod_{i=1}^{N-3} dx_i r^2 dr d\Omega x_i^4 x_j^2 \delta \left(\sum_{i=1}^{N-3} x_i^2 + r^2 - 1 \right)}{\int \prod_{i=1}^{N-3} dx_i r^2 dr d\Omega \delta \left(\sum_{i=1}^{N-3} x_i^2 + r^2 - 1 \right)} \quad \text{for } N \geq 5 \\ &= \frac{\langle x_i^4 x_j^2 x_k^2 \rangle_{N-2}}{\langle x_i^2 \rangle_{N-2}} \quad i \neq j \neq k\end{aligned}$$

so that

$$\langle x_i^4 x_j^2 x_k^2 \rangle_N = \frac{3}{N(N+2)(N+4)(N+6)}$$

which is the highest moment required here. However, it is possible to continue the procedure to obtain all the moments.

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