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Equivalence of population-flow and quantum-mechanical derivations of quadrupolar multiplet intensities

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ABSTRACT

In a previous article (*Concepts Magn. Reson. Part A*, **40**A, 90–99, 2012) a simple explanation for the relative intensities of the components of quadrupolar-split nuclear magnetic resonance (NMR) multiplets was presented. The derivation avoided angular-momentum operator algebra and instead used a population-flow argument based on Boltzmann statistics and relaxation from magnetic saturation. Some colleagues subsequently suggested that the agreement between this intuitive derivation and the standard quantum-mechanical result might be coincidental. In this paper, we show explicitly that the two approaches are mathematically equivalent. The excess-spin construction used previously corresponds directly to the diagonal elements of the deviation density matrix, and the recursive population-flow relations map onto the same quadratic dependence that arises from the matrix elements of the transverse spin operator. Thus, the derivation is neither heuristic, approximate, nor accidental: it is an alternative representation of the same underlying quantum-mechanical structure. The presented work gives an intuitive understanding of the NMR peak intensities observed for quadrupolar nuclear spins embedded in anisotropic materials.

Introduction

Motivation: Anisotropic materials, such as stretched hydrogels or liquid crystals, provide reproducibly aligned macroscopic environments for analytical NMR spectroscopy [1,2] and mimic some aspects of cellular and tissue structure [3]. When quadrupolar nuclei are dissolved in such media, their NMR spectra often display pronounced, well-resolved multiplet splittings since their quadrupolar couplings are no longer averaged to zero. This has been observed for the quadrupolar nuclear spins ^{2}H, ^{7}Li, ^{9}Be, ^{17}O, ^{23}Na, ^{27}Al, ^{95}Mo, and ^{133}Cs., [4] A recent example spectrum is shown in Fig. 1. These experimentally accessible multiplets raise a natural theoretical question: what determines the relative intensities of the component lines?

The simple rule: Quadrupolar nuclei ($I > 1/2$) in anisotropic environments exhibit characteristic multiplets whose peak intensities follow

$$A(j) \propto j(\mathscr{N} - j),\ \mathscr{N} = 2I + 1. \tag{1}$$

Here $j = 1,\ 2, \ldots, \mathscr{N} - 1$ indexes the positions of the allowed single-quantum transitions along the Zeeman ladder. We take $j = 1$ to denote the transition between the two lowest Zeeman levels, and then increase j stepwise upwards through the ladder (towards lower m).

Each transition is single-quantum with magnetic quantum number change $\Delta m = -1$, *i.e.*, $m \rightarrow m - 1$, where m denotes the magnetic quantum number of the initial Zeeman level involved in the transition. The spin quantum number is I, giving $\mathscr{N} = 2I + 1$ Zeeman levels.

In our earlier paper [4] we arrived at this expression without invoking the usual angular-momentum operator formalism. Instead, we used a population-based argument: equalization of energy-level populations under magnetic saturation, followed by relaxation back to the Boltzmann distribution, with the net flow of spins between adjacent levels determining the relative line intensities.

The derivation was deliberately elementary and pedagogical. Its purpose was to show that the intensity pattern arises from simple physical considerations. However, the absence of explicit quantum-

Abbreviations: $A(j)$, intensity of the j-th transition; CW, continuous wave; I, spin quantum number; I_x, x-angular momentum operator; I_y, y-angular momentum operator; $\mathscr{N} = 2I + 1$, number of Zeeman levels; N_i, excess population in level i (intuitive model); ρ_{mm}, diagonal density-matrix element; $\Delta\rho_{mm}$, deviation density-matrix element; RF, radio frequency; S_i, net spin flow between levels i and $i + 1$.

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mechanical operators led some colleagues to suggest that the agreement with the standard result might be coincidental.

The aim of the present paper is to dispel this concern. Our purpose is not to re-establish the well-known equivalence of continuous wave (CW) and pulsed NMR, which is treated in standard texts such as [5]. We show that the population-flow argument is formally equivalent to the density-matrix derivation used in quantum mechanics [5–7] and in particular the apparently heuristic recursion used in the intuitive model produces exactly the same quadratic factor that arises from the matrix element $\langle I,m|I_x|I,m-1\rangle$. This explicit algebraic identification has not previously been shown. The two approaches differ only in language, not in substance.

Summary of the population-flow derivation

Equilibrium populations: We have $\mathscr{N} = 2I+1$ equally spaced Zeeman energy levels, indexed from the bottom by $i = 1,\ 2, \ldots, \mathscr{N}$. In the high-temperature limit, the equilibrium population can be defined as

$$N_i^{\mathrm{eq}} = 2(\mathscr{N} - (i-1)), \tag{2}$$

with $i = 1$: $N_1^{\mathrm{eq}} = 2\mathscr{N}$ (bottom level, largest population) and $i = \mathscr{N}$: $N_{\mathscr{N}}^{\mathrm{eq}} = 2$ (top level, lowest population). The sequence is linear in i, decreasing by 2 each step. Eq. (2) is valid in the high-temperature limit, where the Boltzmann distribution is also linear. The factor of 2 is arbitrary and serves only to produce an integer-valued linear sequence; any constant scaling would yield the same relative intensities. It proves convenient as it ensures all N_i^{eq}, N_i^{sat} and ΔN_i are integers regardless of spin I.

Total equilibrium population: The total equilibrium population is the sum over all the Zeeman levels

$$\sum_{i=1}^{\mathscr{N}} N_i^{\mathrm{eq}} = 2\sum_{i=1}^{\mathscr{N}} (\mathscr{N} - i + 1) = 2\sum_{i=1}^{\mathscr{N}} (\mathscr{N}+1) - 2\sum_{i=1}^{\mathscr{N}} i. \tag{3}$$

Therefore,

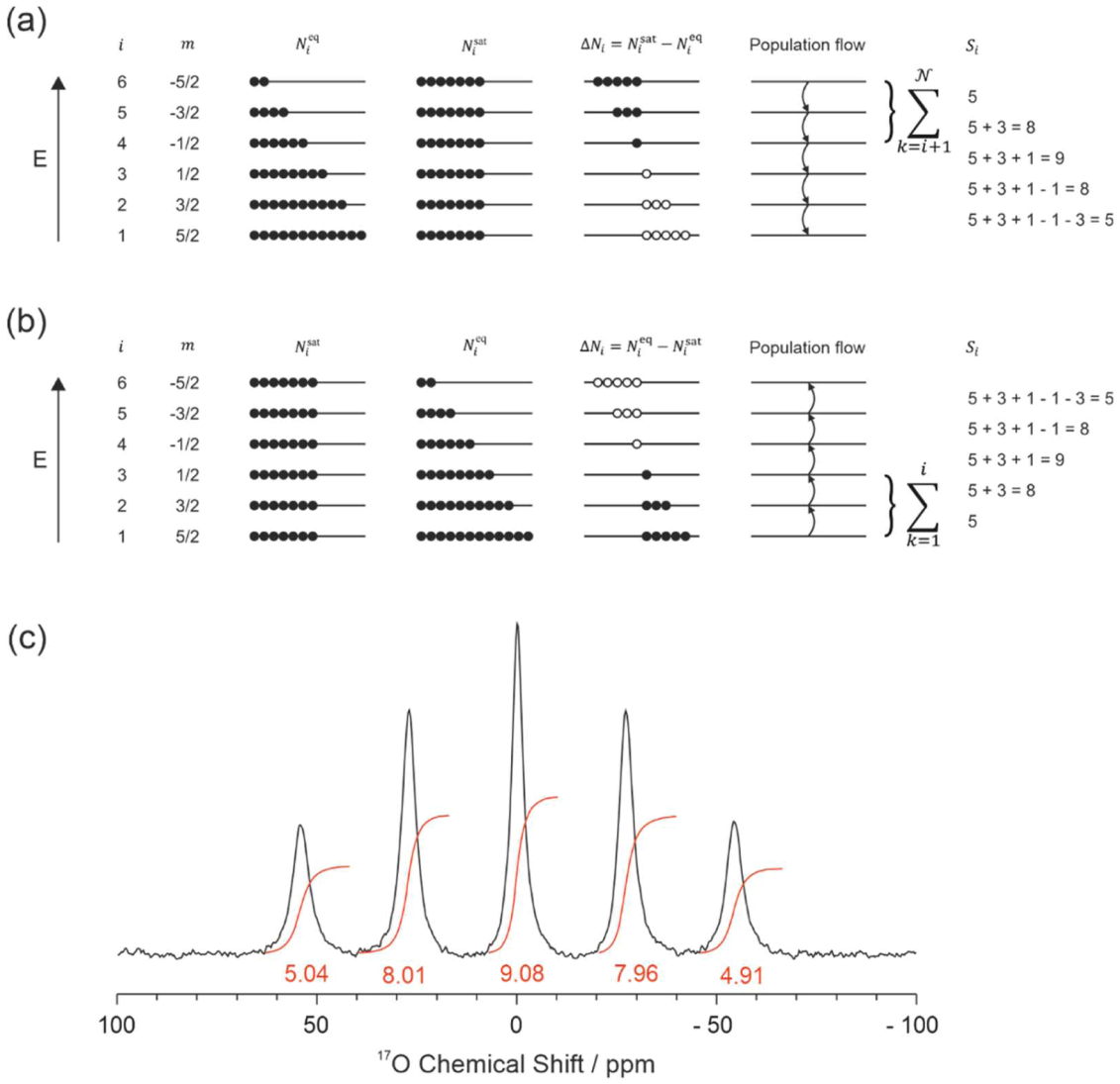

Fig. 1. Schematic energy (E) level diagrams for a spin $I = 5/2$ nucleus showing the distribution of populations at equilibrium N_i^{eq}, at saturation N_i^{sat} and the excess population ΔN_i defined in (a) for the top-down recursion of spin flow, and in (b) the bottom-up recursion of spin flow. The relative peak intensities in the spectrum are given by the cumulative sum (a) from level $i+1$ to N and (b) from level 1 to i. The Zeeman levels are ordered from bottom ($i = 1$) to top ($i = \mathscr{N}$) with magnetic quantum numbers $m = I,\ I-1,\ \ldots,\ -I$. Filled and open symbols correspond to positive and negative excess populations. (c) Relevant portion of the natural abundance ^{17}O NMR spectrum from an aqueous SDS/decanol liquid crystal sample [8] acquired at 9.4 T and 298 K with 16,384 transients using a standard 90° excitation pulse followed by acquisition of the free induction decay. The relative integrals are in red, and the residual quadrupolar coupling is 1.47 kHz.

$$\sum_{i=1}^{\mathscr{N}} N_i^{\mathrm{eq}} = \mathscr{N}(\mathscr{N}+1). \tag{4}$$

Saturated populations: Under complete magnetic saturation, the excess population is evenly distributed and is therefore the total equilibrium population divided by the number of states. Using Eq. (4)

$$N_i^{\mathrm{sat}} = \frac{\mathscr{N}(\mathscr{N}+1)}{\mathscr{N}} = \mathscr{N}+1. \tag{5}$$

Recursion for spin flow: The redistribution of populations between Zeeman levels may be described either as relaxation from saturation back to equilibrium or as excitation from equilibrium towards saturation. These are complementary bookkeeping descriptions of the same physical process: (*i*) a top-down formulation, Fig. 1(a), in which spin flow is accumulated downwards from the highest level, and (*ii*) a bottom-up formulation, Fig. 1(b), in which spin flow is accumulated upwards from the lowest level.

Top-down formulation: In the earlier paper [4] the top-down formulation of population flow was used to arrive at the intuitive result. The following is the formal derivation of that result.

Let S_i be the net spin flow between levels i and $i+1$ during redistribution of spins from the saturated state back to equilibrium, with downward flow from $i+1$ to i taken as positive. The quantity S_i is therefore defined for $i = 1, 2, \ldots, \mathscr{N}-1$. At the top of the Zeeman ladder there is no level above $\mathscr{N}$. We formally define $S_{\mathscr{N}} = 0$ to represent the absence of inflow above level $\mathscr{N}$.

Defining the excess population as the deviation of the saturated population from the equilibrium population gives,

$$\Delta N_i = N_i^{\mathrm{sat}} - N_i^{\mathrm{eq}} = 2i - \mathscr{N} - 1. \tag{6}$$

This sequence is linear in i, increasing by 2 with each step. With $i = 1$: $\Delta N_i = 1 - \mathscr{N}$ (bottom level, population excess is negative because the equilibrium population is greater than the saturated population), and for $i = \mathscr{N}$: $\Delta N_i = \mathscr{N} - 1$ (top level, population excess is positive because the saturated population exceeds the equilibrium population). This function is symmetric about $i = (\mathscr{N}+1)/2$, with zero excess at this level, positive population excess above it and negative population excess below it. The sum of excess populations across all levels is therefore

$$\sum_{i=1}^{\mathscr{N}} \Delta N_i = 0. \tag{7}$$

Since S_i represents inflow from level $i+1$ and S_{i-1} represents outflow to level $i-1$, conservation of population for level i gives

$$S_{i-1} - S_i = \Delta N_i, \tag{8}$$

Using the top boundary condition $S_{\mathscr{N}} = 0$, we therefore sum downwards. The net spin flow between levels $i+1$ and i is equal to the total excess population in all levels above that transition:

$$S_i = \sum_{k=i+1}^{\mathscr{N}} \Delta N_k = \sum_{k=i+1}^{\mathscr{N}} (2k - \mathscr{N} - 1). \tag{9}$$

Evaluating the sum gives

$$S_i = 2\sum_{k=i+1}^{\mathscr{N}} k - \sum_{k=i+1}^{\mathscr{N}} (\mathscr{N}+1), \tag{10}$$

and hence

$$S_i = \mathscr{N}(\mathscr{N}+1) - i(i+1) - (\mathscr{N}-i)(\mathscr{N}+1). \tag{11}$$

Therefore,

$$S_i = i(\mathscr{N}-i). \tag{12}$$

Bottom-up formulation: Equivalently, we may describe the same redistribution starting from the bottom of the Zeeman ladder. In this case, we define the excess population as the deviation of the equilibrium population from the saturated population,

$$\Delta N_i = N_i^{eq} - N_i^{sat} = \mathscr{N} + 1 - 2i. \tag{13}$$

which is the negative of the top-down definition (Eq. (6)).

We now define upward flow as positive and therefore conservation of population for level i gives

$$S_i - S_{i-1} = \Delta N_i, \tag{14}$$

since S_{i-1} represents inflow from level $i-1$ to level i, and S_i represents outflow from level i to level $i+1$.

At the bottom of the Zeeman ladder there is no level below level $i = 1$, so the boundary condition is $S_0 = 0$. Iterating from the lower boundary yields

$$S_i = \sum_{k=1}^{i} \Delta N_k, \tag{15}$$

and substitution of $\Delta N_k = \mathscr{N} + 1 - 2k$ gives the same result as the top-down formulation

$$S_i = i(\mathscr{N}-i). \tag{16}$$

In the population picture, the deviations $N_i^{\mathrm{eq}} - N_i^{\mathrm{sat}}$ form a linear sequence in the level index i. The cumulative spin flow S_i is the discrete integral of this linear sequence, which produces a second-degree (quadratic) dependence on i.

We emphasise that magnetic saturation is introduced as a conceptual device rather than an experimental requirement (the Appendix contains an extension of this idea). The population-flow model uses the difference between equilibrium and saturated populations because this difference forms a strictly linear sequence in the level index i.

Transition intensities: The two approaches differ only in sign convention and direction of accumulation. Both formulations therefore compute the same cumulative population imbalance between each level. Since the intensity $A(j)$ of the j-th transition is proportional to the net spin flow between adjacent levels, we obtain

$$A(j) \propto S_j = j(\mathscr{N}-j). \tag{17}$$

independent of the chosen formulation. Because the recursion is linear and the total excess population sums to zero, both approaches lead to the identical result since transverse magnetization corresponds to the superposition of upward and downward transitions, which have the same amplitudes.

Density matrix formalism

In the quantum-mechanical description of NMR, the state of a spin ensemble is represented by the density matrix, whose diagonal elements give the populations of the Zeeman levels, and the off-diagonal elements describe coherences generated during excitation and detection.

At thermal equilibrium, the diagonal elements form a weakly tilted linear ramp in the magnetic quantum number m, reflecting the Boltzmann distribution in the high-temperature limit. Thus,

$$\rho_{mm}^{\mathrm{eq}} \propto N_m^{\mathrm{eq}}, \tag{18}$$

since the diagonal elements of the density matrix are proportional to the populations of the corresponding Zeeman levels.

Magnetic saturation, by contrast, drives all populations toward equality. The deviation density matrix is therefore defined as

$$\Delta\rho_{mm} = \rho_{mm}^{\mathrm{eq}} - \rho_{mm}^{\mathrm{sat}}, \tag{19}$$

which measures how far the population of each level deviates from its saturated value. This is the quantum-mechanical analogue of the "excess spins" ΔN_i introduced in the population flow model. In both descriptions the deviations form a linear sequence across the Zeeman ladder that sum

to zero, differing only by an overall proportionality constant.

In the quantum mechanical treatment, the intensity of the $m \rightarrow m-1$ transition is proportional to the product of the population difference between the two levels and the square of the corresponding matrix element of the transverse spin operator [6]

$$A_m^{\mathrm{QM}} \propto \left(\rho_{m-1,m-1}^{\mathrm{eq}} - \rho_{mm}^{\mathrm{eq}}\right) |\langle I, m \,|\, I_x \,|\, I, m-1\rangle|^2. \tag{20}$$

This expression introduces the operator formalism, but it does not change the underlying physical picture: the population difference appearing in Eq. (20) simply reflects the equilibrium population gradient across the Zeeman ladder.

In the high-temperature approximation, the equilibrium diagonal elements vary approximately linearly with the magnetic quantum number m,

$$\rho_{mm}^{\mathrm{eq}} \propto 1 + \epsilon\, m, \tag{21}$$

where $\epsilon = \hbar\gamma B_0/k_{\mathrm{B}}T \ll 1$, $\hbar$ is the reduced Planck's constant, γ is the magnetogyric ratio, B_0 is the magnetic induction, k_{B} is the Boltzmann constant and T is the absolute temperature.

From Eqs. (19) and (21), we can write:

$$\Delta\rho_{mm} - \Delta\rho_{m-1m-1} = \rho_{mm}^{\mathrm{eq}} - \rho_{m-1m-1}^{\mathrm{eq}} = \epsilon, \tag{22}$$

Since

$$\rho_{mm}^{\mathrm{sat}} - \rho_{m-1m-1}^{\mathrm{sat}} = 0. \tag{23}$$

Hence the population differences $\rho_{m-1,m-1}^{\mathrm{eq}} - \rho_{mm}^{\mathrm{eq}}$ are approximately constant across the multiplet, mirroring the linear variation of N_i^{eq} used in the population-flow derivation. The result implies that the population difference between adjacent energy levels is identical. This comparison is only possible in the high-temperature limit, otherwise the ramp of 'excess spins' would be non-linear.

Matrix elements: In the quantum-mechanical description, a spectral line appears because the radio frequency (RF) field induces transitions between adjacent Zeeman levels. The strength of such a transition is determined by how strongly the RF Hamiltonian couples the two states involved. For a linearly oscillating RF field, the relevant part of the Hamiltonian is proportional to the transverse spin operator. Depending on the phase of the RF pulse, the transverse operator may be I_x or I_y; both yield identical transition intensities and the choice of I_x is simply conventional. Thus, the quantity that determines the efficiency of the $m \rightarrow m-1$ transition is the transition matrix element of I_x between those two states.

A transition matrix element describes the overlap between the initial and final states after the operator has acted on the initial state. In this case, the operator I_x connects neighbouring Zeeman levels, and the magnitude of $\langle I, m \,|\, I_x \,|\, I, m-1\rangle$ quantifies how strongly the RF field mixes those two states. The larger this overlap, the more transverse magnetization is produced, and the more intense the corresponding spectral line.

For angular-momentum eigenstates, this matrix element has the standard form [6]

$$\langle I, m \,|\, I_x \,|\, I, m-1\rangle = \frac{1}{2}\sqrt{I(I+1) - m(m-1)}. \tag{24}$$

Since the intensity is proportional to the square of this quantity, the quantum-mechanical expression becomes

$$A_m^{\mathrm{QM}} \propto I(I+1) - m(m-1), \tag{25}$$

which is a quadratic function in the magnetic quantum number m. This quadratic dependence mirrors the second-order structure that emerged from the population-flow recursion, as we show next.

Equivalence of the two expressions: To compare the quantum-mechanical expression with the population-flow result, we express the transition j in terms of I and m.

$$j = I - m + 1, \tag{26}$$

which labels the transitions from the bottom of the Zeeman ladder upwards, exactly as in the population-flow derivation.

Substituting $m = I - j + 1$ into the quadratic factor in Eq. (25) gives

$$I(I+1) - m(m-1) = I(I+1) - (I-j+1)(I-j). \tag{27}$$

Which simplifies to

$$I(I+1) - (I-j+1)(I-j) = j(2I+1-j). \tag{28}$$

Since the number of Zeeman levels is $\mathscr{N} = 2I+1$, we obtain

$$I(I+1) - m(m-1) = j(\mathscr{N} - j). \tag{29}$$

Thus, the quantum-mechanical intensity is

$$A_{\mathrm{QM}}(j) \propto j(\mathscr{N} - j). \tag{30}$$

This is identical to the population-flow expression obtained in Eq. (17). The two derivations are not merely consistent; they are algebraically equivalent. The matrix element $\langle I, m|I_x|I, m-1\rangle$ carries the factor $\sqrt{I(I+1) - m(m-1)}$ whose squared modulus is a quadratic polynomial in m. After relabelling m in terms of j, this becomes the same $j(\mathscr{N} - j)$ dependence obtained from the population-flow recursion.

Conclusions

The agreement between the population-flow and the density matrix derivations reflects their shared physical structure: linear population differences, conservation of total population and single-quantum transitions between adjacent Zeeman levels. This understanding is key to the appearance of NMR spectra from quadrupolar nuclear spins in aligned materials such as stretched hydrogels, liquid crystals and other oriented media. While the population flow model gives an intuitive understanding of the peak intensities, several quantum mechanical properties are clearly not predicted by this simple model. In the quantum mechanical treatment, the transverse operator $I_x = 1/2(I_+ + I_-)$ couples adjacent Zeeman levels through the raising and lowering operators I_+ and I_-, which are Hermitian conjugates. The population-flow model does not distinguish between these operators individually; it captures only the net redistribution of populations between adjacent levels that determine the relative transition intensities, but not the phase or direction of the underlying transitions. A complete description of quadrupolar spin dynamics still requires the full density matrix formalism, including spherical tensor operators and the explicit Hamiltonians governing the system.

We reiterate that the present analysis does not rederive the general equivalence of CW and pulsed NMR spectra, which is well established. Instead, it demonstrates that the specific population-flow construction introduced previously is algebraically identical to the standard density-matrix expression from transition intensities.

A further point of potential ambiguity remains: does our assumption of magnetic saturation compromise any of the foregoing conclusions? The answer is No, and for completeness the reasoning behind this is set out in the Appendix.

CRediT authorship contribution statement

Philip W. Kuchel: Writing – review & editing, Writing – original draft, Conceptualization. **Stuart J. Elliott:** Writing – review & editing. **Thomas R. Eykyn:** Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix

Partial magnetic saturation

Saturation is used in the main text as a mathematical device to generate a linear population-deviation profile; it is not required experimentally, as is shown next.

Population-flow description: To represent uniform partial saturation, we introduce a parameter $0 \leq \lambda \leq 1$ describing the extent of saturation. For $\lambda = 0$ the system is at equilibrium; for $\lambda = 1$ it is fully saturated. Under partial saturation the energy-level populations may be written as a linear interpolation between their equilibrium and fully saturated values,

$$N_i^{(\lambda)} = N_i^{eq} + \lambda\left(N_i^{sat} - N_i^{eq}\right). \tag{A1}$$

The corresponding excess population becomes

$$\Delta N_i^{(\lambda)} = N_i^{eq} - N_i^{(\lambda)} = \lambda\left(N_i^{eq} - N_i^{sat}\right) = \lambda \Delta N_i. \tag{A2}$$

Thus, partial saturation simply rescales the excess population sequence by λ while preserving its linear form.

Let $S_i^{(\lambda)}$ denote the cumulative spin flow from level i to $i+1$. Because the population-flow recursion is linear in the excess populations ΔN_i, the resulting spin flow scales identically,

$$S_i^{(\lambda)} = \lambda S_i = \lambda\, i(\mathscr{N} - i). \tag{A3}$$

Since $S_i = i(\mathscr{N} - i)$ for complete saturation, Eq. (16), the population flow under partial saturation simply rescales the spin flow by a constant factor λ without altering its characteristic $i(\mathscr{N} - i)$ dependence. The relative peak intensities therefore remain unchanged.

Correspondence with the density matrix description: In the quantum mechanical formalism, the deviation density matrix is

$$\Delta\rho_{mm} = \rho_{mm}^{eq} - \rho_{mm}^{sat}. \tag{A4}$$

Under partial saturation the diagonal elements is similarly an interpolation of equilibrium and saturation values,

$$\rho_{mm}^{(\lambda)} = \rho_{mm}^{eq} + \lambda\left(\rho_{mm}^{sat} - \rho_{mm}^{eq}\right). \tag{A5}$$

Therefore

$$\Delta\rho_{mm}^{(\lambda)} = \rho_{mm}^{eq} - \rho_{mm}^{(\lambda)} = \lambda\left(\rho_{mm}^{eq} - \rho_{mm}^{sat}\right) = \lambda\Delta\rho_{mm}. \tag{A6}$$

Thus, the deviation density matrix scales by the same factor λ.

Because the Liouville–von Neumann equation governing the density matrix is linear in ρ, the resulting transition intensities scale identically. Using Eq. (17) from the main text, this becomes

$$A_{QM}^{(\lambda)} \propto \lambda\, j(\mathscr{N} - j). \tag{A7}$$

Uniform partial saturation therefore multiplies all spectral lines by the same factor while preserving their relative intensities. Thus, both the population-flow model and the density matrix formalism predict that partial magnetic saturation scales all multiplet intensities uniformly while leaving their characteristic $j(\mathscr{N} - j)$ pattern unchanged.

Data availability

Data will be made available on request.

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