THEORY OF COHERENT MULTI-QUANTUM NMR IN MULTILEVEL SPIN SYSTEM

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Abstract When spin system absorbs the rf radiation of strong intensity coherent transitions due to the simultaneous absorbtion of many photons can be observed[1,2]. In this article we discuss a new approach to the description of multi-photon coherent transition in which the spin Hamiltonian is formulated in terms of the standard-basis operators[3] and the technique of coherent averaging is employed.

The substitution of the spin operators by standard-basis operators called also the population operators[4] is naturaly adapted to systems having nonequidistant energy levels in which these operators are represented by matrices whose elements are zero except for single one. They provide a clear picture of interactions which cause inter-level transitions obscured by the use of fictitious angular momentum operators. The diagonal operator N_m plays the role of the occupation number operator for individual spin level while the nondiagonal N_m^+ and N_m^- generate the transitions $m \to m+1$ and $m \to m-1$ respectively. Thus the Hamiltonian of the quadrupole perturbed spin system irradiated by the rf field of frequency ω is expressed as

$$H(t) = H_q + H_{rf}(t) = \sum_{m=-s}^{m=s} [E_m N_m + 2h\omega_1 A_m (N_m^+ + N_{m+1}^-) \cos \omega t] \qquad (1)$$

with $E_m = -\hbar\omega_0 m + \frac{1}{3}\hbar\omega_q[3m^2 - s(s+1)]$ and $A_m = \sqrt{s(s+1) - m(m+1)}$.

The time evolution operator for this system might be separated into the part due to the static field alone and the part representing the effect of the rf field. The time dependence of $H_{rf}(t)$ requires time ordering which prevents to actually perform the integration of the exponent of the time-evolution operator. To overcome this the coherent averaging is used and the exponent of the rf part is expanded as:

$$U(t) = \exp(-i H_q t).\exp(-i \int^t [H_{0rf}(t') + H_{1rf}(t') + H_{2rf}(t') + \dots] dt' \qquad (2)$$

By substituting (1) into (2) the effective rf Hamiltonian is obtained which in the zeroth-order approximation $H_{0rf}(t)$ gives one-photon transitions, the first two terms of the first-order approximation

$$H_{1rf}(t) = \frac{1}{2}\frac{\omega_1^2}{Q}\sum_m [b_m(t) N_{m+1}^+ N_m^+ + b_m^* N_{m+1}^- N_{m+2}^- + 2c_m(t) N_m(t)] \qquad (3)$$

generate the two-photon transitions between $m \rightleftarrows m+2$ levels while the last term induces Bloch-Siegert shift, and the second-order approximation

$$H_{2rf}(t) = \frac{i}{6}\frac{\omega_1^3}{Q^2}\sum_m [d_m(t) N_m^+ + d_m^*(t) N_{m+1}^- + e_m(t) N_{m+2}^+ N_{m+1}^+ N_m^+ + e_m^* N_{m+1}^- N_{m+2}^- N_{m+3}^-] \qquad (4)$$

gives the three-photon transitions $m \rightleftarrows m+3$ induced by the last two terms. Further expansion of the series (2) which is convergent for $\omega_1 < Q$ gives the other multi-photon transitions.

Detail calculation of the coeficient b,c,d and e reveals a features[5] not obtained in the previous considerations[2]. For example, $H_{1rf}(t)$ induces not only the usual double-photon transitions[2] when $\hbar\omega = \frac{1}{2}(E_{m+2} - E_m)$ but also when $\hbar\omega = E_{m+1} - E_m$, and $\hbar\omega = E_{m+2} - E_{m-1}$.

This formalism can be also applied when treating the optical multi-photon coherence.

References

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