

NUCLEAR MAGNETIC RELAXATION BY MULTIPOLE INTERACTIONS IN LIQUIDS

BY J. S. Blicharski

Institute of Physics, Jagellonian University, Cracow*

Institute of Nuclear Physics, Cracow

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Transition probabilities and spin-lattice relaxation times have been calculated in the presence of multipole interactions of arbitrary order in liquids.

Consider a single nucleus of an arbitrary spin I in a molecule in liquid. The Hamiltonian \mathcal{H} of intramolecular interaction between the nucleus and electronic cloud [1] can be presented as the sum of electric and magnetic multipole interactions $\hbar G^{(l)}$ of order 2^l

$$\mathcal{H} \equiv \hbar G = \hbar \sum_{l=0}^{2I} G^{(l)} \quad (1)$$

where the electric and magnetic multipole interactions usually appear for even and odd number l respectively. The arbitrary multipole interaction $\hbar G^{(l)}$ may be expressed as a scalar product of two spherical tensors A_{lm} and F_{lm} , dependent on the nuclear coordinates r_n and electron coordinates r_e respectively

$$\hbar G^{(l)} = \sum_{m=-l}^l (-1)^m A_{lm} F_{l-m} \quad (2)$$

In the case of liquids, the orientation of a molecule is a random function of time because of rotational diffusion and, as a consequence, the elements of the tensor $F_{lm} = F_{lm}(t)$ are random functions of time. In the presence of a strong external magnetic field H_0 the multipole interactions may be treated as small time-dependent perturbations. These perturbations can produce the transitions between different Zeman states $|\alpha\rangle = |Im_\alpha\rangle$ and $|\beta\rangle = |Im_\beta\rangle$. According to the perturbation theory the probability $W_{\alpha\beta}$ of the transitions per unit of time may be expressed as spectral density of the correlation function

$$W_{\alpha\beta} = \int_{-\infty}^{+\infty} \langle G_{\alpha\beta}(0) G_{\alpha\beta}^*(\tau) \rangle e^{i\omega_{\alpha\beta}\tau} d\tau \quad (3)$$

* Address: Instytut Fizyki, Uniwersytet Jagielloński, Kraków, Reymonta 4, Poland.

with

$$\langle G_{\alpha\beta}(0)G_{\alpha\beta}^*(\tau) \rangle = \langle |G_{\alpha\beta}|^2 \rangle e^{-\frac{|\tau|}{\tau_c}} \quad (4)$$

$$G_{\alpha\beta}(t) = (\alpha|G(t)|\beta) = \sum_{lm} (-1)^m (\alpha|A_{lm}|\beta) F_{l-m}(t) \quad (5)$$

$$\omega_{\alpha\beta} = (\alpha - \beta)\omega_0 \equiv (m_\alpha - m_\beta)\omega_0 \quad (6)$$

where ω_0 is Larmor precession angular frequency for the considered nucleus in the H_0 field and τ_c is the molecular correlation time.

The matrix elements $(\alpha|A_{lm}|\beta)$ of the tensor operator A_{lm} may be expressed in the form

$$(\alpha|A_{lm}|\beta) = (-1)^{l+I+\alpha} M_n^{(l)} \frac{\sqrt{(2I-l)!(2I+l+1)!}}{(2I)!} \begin{pmatrix} I & I & l \\ -\alpha & \beta & m \end{pmatrix} \quad (7)$$

$$M_n^{(l)} \equiv (II|A_{l0}|II) \quad (8)$$

where $\begin{pmatrix} I_1 & I_2 & I_3 \\ m_1 & m_2 & m_3 \end{pmatrix}$ are Wigner's 3j symbols [2] and $M_n^{(l)}$ are nuclear multipole moments equal to $Ze, \mu, eQ/2, -\Omega, Q^{(4)}$ for $l = 0, 1, 2, 3, 4$ respectively [1], [3].

Taking into account the orthogonality of the functions $F_{lm}(t)$ and assuming the isotropic molecular motion, one finds

$$W_{\alpha\beta} = \sum_{lm} |(\alpha|A_{lm}|\beta)|^2 \langle |F_{lm}|^2 \rangle j(\omega_{\alpha\beta}) \quad (9)$$

with

$$\langle |F_{lm}|^2 \rangle = \frac{\sum_m |F_{lm}^0|^2}{2l+1} \quad (10)$$

$$j(\omega) = \frac{2\tau_c}{1 + \omega^2 \tau_c^2} \quad (11)$$

where F_{lm}^0 are constant values of the function $F_{lm}(t)$ in the molecular reference frame. In particular, the values $F_{l0}^0 \equiv M_e^{(l)}$ may be called the multipole moments of the electronic shell. These values are equal to

$$V, H_z, \frac{1}{2} \frac{\partial^2 V}{\partial z^2} \equiv \frac{1}{2} eq, \frac{1}{3!} \frac{\partial^2 H_z}{\partial z^2}, \frac{1}{4!} \frac{\partial^4 V}{\partial z^4}$$

for $l = 0, 1, 2, 3, 4$ respectively where V and H are the electric potential and local magnetic field produced by the electronic cloud at the position of the considered nucleus.

Finally, using the Eqs (3)–(11) one can present the transition probability $W_{\alpha\beta}$ in the following form

$$W_{\alpha\beta} = \sum_l W_{\alpha\beta}^{(l)} \quad (12)$$

$$W_{\alpha\beta}^{(l)} = \langle |G_{\alpha\beta}|^2 \rangle j(\omega_{\alpha\beta}) = \frac{c_l^2 (1+a_l) (2l-1)! (2l+1)!}{(2l+1) [(2l)!]^2} \times \\ \times \begin{pmatrix} l & l & l \\ -\alpha & \beta & \alpha - \beta \end{pmatrix}^2 j(\omega_{\alpha\beta}) \quad (13)$$

$$\hbar c_l \equiv M_n^{(l)} M_e^{(l)} \quad (14)$$

$$a_l \equiv \frac{\sum_{m \neq 0} |F_{lm}^0|^2}{(M_e^{(l)})^2} \quad (15)$$

where c_l is the coupling constant for multipole interaction and a_l may be called the asymmetry factor.

The above-presented matrix elements and transition probabilities may also be calculated directly if one uses an effective spin Hamiltonian for multipole interactions. This Hamiltonian can be constructed from Eq. (2) by introducing the nuclear spin components I_x, I_y, I_z in the place of the position variables x_n, y_n, z_n , followed by symmetrization. The explicit form of the spin Hamiltonian is well known for magnetic dipole and electric quadrupole interaction [1]. In the Appendix of this paper the effective spin Hamiltonian is presented for magnetic octupole and electric hexadecapole interaction.

The time dependence of the expectation value $\langle I \rangle(t)$ of the nuclear spin components $I = (I_x, I_y, I_z)$ in the presence of multipole interactions may be calculated on the basis of the theory of the density matrix $\sigma_{\alpha\alpha'}$, [4], [5]

$$\langle I \rangle(t) = \sum_{\alpha\alpha'} I_{\alpha'\alpha} \sigma_{\alpha\alpha'}(t) \quad (16)$$

$$\frac{d\sigma_{\alpha\alpha'}}{dt} + i\omega_{\alpha\alpha'} \sigma_{\alpha\alpha'} = \sum_{\beta\beta'} R_{\alpha\alpha'\beta\beta'} (\sigma_{\beta\beta'} - \sigma_{\beta\beta'}^T) \quad (17)$$

where $\sigma_{\alpha\alpha'}^T$ are density matrix elements at the thermal equilibrium and $R_{\alpha\alpha'\beta\beta'}$ is the relaxation matrix.

Further consideration will be limited to the longitudinal spin component I_z (along H_0 field) obeying the eigenequation

$$I_z |\alpha\rangle = m_\alpha |\alpha\rangle \equiv \alpha |\alpha\rangle. \quad (18)$$

In this case one can find

$$\langle I_z \rangle(t) = \sum_\alpha \alpha p_\alpha(t) \quad (19)$$

$$\frac{dp_\alpha}{dt} = \sum_\alpha R_{\alpha\beta}(p_\beta - p_\beta^T) \quad (20)$$

$$p_\alpha = \sigma_{\alpha\alpha} \quad (21)$$

$$R_{\alpha\beta} \equiv R_{\alpha\alpha\beta\beta} = W_{\alpha\beta} - \delta_{\alpha\beta} \sum_\gamma W_{\alpha\beta} \quad (22)$$

where p_α are the relative populations of the Zeman levels and $\delta_{\alpha\beta}$ is Kronecker's delta. The rate of change of the expectation value $\langle I_z \rangle$ may be written in the form

$$\frac{d\langle I_z \rangle}{dt} = \sum_{\alpha\beta} \alpha R_{\alpha\beta} p_\beta \quad (23)$$

In the extreme narrowing case for $\omega_0\tau_c \ll 1$ one can note that the sum $\sum_\alpha \frac{\alpha}{\beta} R_{\alpha\beta}$ is independent of β . Taking into account the above fact and the definition of the spin-lattice relaxation time T_1 , one can present Eq. (23) in the form

$$\frac{d\langle I_z \rangle}{dt} = -\frac{1}{T_1} (\langle I_z \rangle - I_0) \quad (24)$$

$$\frac{1}{T_1} = -\sum_\alpha \frac{\alpha}{\beta} R_{\alpha\beta} = \sum_\alpha \left(\frac{\beta - \alpha}{\beta} \right) W_{\alpha\beta} \quad (25)$$

with solution

$$\frac{\langle I_z \rangle(t) - I_0}{\langle I_z \rangle(0) - I_0} \equiv R(t) = e^{-\frac{t}{T_1}} \quad (26)$$

where $R(t)$ is the relaxation function and I_0 is the value of $\langle I_z \rangle$ at the thermal equilibrium.

As a final result, for the relaxation time T_1 , it follows from the Eqs (11)–(13) and (25) that:

$$\frac{1}{T_1} = \sum_l \left(\frac{1}{T_1} \right)_l \quad (27)$$

$$\begin{aligned} \left(\frac{1}{T_1} \right)_l &= \frac{2c_l^2(1+a_l)\tau_c}{(2l+1)I(2l)!} \sum_{n=1}^l \frac{(2l-n)!(l+n)!}{(n-1)!(l-n)!} = \\ &= \frac{c_l^2(1+a_l)l(l+1)\tau_c}{(2l+1)I(I+1)} \prod_{n=1}^l \left(\frac{2I+n+1}{2I-n+1} \right). \end{aligned} \quad (28)$$

The above-presented results for T_1 may also be derived on the basis of the well-known Gorter relation [6]

$$\frac{1}{T_1} = \frac{1}{2} \frac{\sum_{\alpha\beta} (\alpha-\beta)^2 W_{\alpha\beta}}{\sum_{\alpha} \alpha^2} = \frac{3}{2} \frac{\sum_{\alpha\beta} (\alpha-\beta)^2 W_{\alpha\beta}}{I(I+1)(2I+1)} \quad (29)$$

The application of the Eqs (11)–(13) and (29) in the extreme narrowing case leads to the same final result as that presented in Eqs (27) and (28). Moreover, in this case ($\omega_0\tau_c \ll 1$) one can expect to find the spin-spin relaxation time T_2 equal to T_1 . Thus the Eqs (27) and (28) are valid both for T_1 and T_2 .

As a special case ($l = 1$) one can get from Eq. (28) the following expressions for nuclear relaxation times in the presence of magnetic dipole (M1) interaction and hypothetical electric dipole (E1) interaction [1], [7]

$$\left(\frac{1}{T_1}\right)_{l=1} = \frac{2}{3} c_{\text{eff}}^2 \tau_c \quad (30)$$

$$c_{\text{eff}}^2 = \frac{c_1^2(1+a_1)}{I^2} = \begin{cases} \gamma^2 \langle H^2 \rangle \equiv \left(\frac{\mu}{I\hbar}\right)^2 \langle H^2 \rangle & \text{(for M1)} \\ \gamma_E^2 \langle E^2 \rangle \equiv \left(\frac{\mu_E}{I\hbar}\right)^2 \langle E^2 \rangle & \text{(for E1)} \end{cases} \quad (31)$$

where μ , μ_E , γ , γ_E , $\langle H^2 \rangle$, $\langle E^2 \rangle$ are nuclear magnetic and electric moment, gyromagnetic and gyroelectric ratio, average square of the local magnetic and electric field respectively. The Eqs (30) and (31) may also be applied if the local magnetic field is produced by neighbouring nuclei of spins I_k with gyromagnetic ratios γ_k . A simple calculation leads to the following expression for the effective square of local magnetic field:

$$\langle H^2 \rangle = 2\hbar^2 \sum_k \frac{I_k(I_k+1)\gamma_k^2}{r_{ik}^6} \quad (33)$$

where r_{ik} is the distance between the considered nucleus i and the nucleus k . The T_1 resulting from the Eqs (30), (31) and (33) is identical with that for dipolar relaxation in the system of unlike spins, presented elsewhere [8].

The expressions (30) and (32) for electric dipole relaxation rate $(1/T_1)_{E1}$ may be applied if the local electric field E produced by the electronic cloud is modulated by rapid molecular reorientations in liquids. Generally the electric dipole contribution may be written in the form

$$\left(\frac{1}{T_1}\right)_{E1} = \frac{1}{3} \gamma_E^2 \langle E^2 \rangle j(\omega) \quad (34)$$

In the case of gases the local electric field may be produced during the molecular collisions. A pulse of the local electric field in a collision is proportional to the momentum transfer. It was shown elsewhere [9] that these pulses cannot be independent and in conse-

quence the spectral density $j(\omega)$ has to be modified ($j(0) = 0$). As a final result for electric dipole relaxation rate in monoatomic gases [9–11] one gets:

$$\left(\frac{1}{T_1}\right)_{E1} = \frac{2MkT}{Z^2 e^2} \left(\frac{\mu_E}{I\hbar}\right)^2 \frac{\omega_0^2 \tau_c}{1 + \omega_0^2 \tau_c^2} \quad (35)$$

where the correlation time τ_c is equal to the average time between atomic collisions, M is atomic mass, Z is atomic number, k is Boltzmann constant and T is absolute temperature:

In the case of crystals of density ρ the calculations [9], [12] lead to the following result:

$$\left(\frac{1}{T_1}\right)_{E1} = \frac{M^2 k T \omega_0^4}{2\pi^2 Z^2 e^2 \rho c^3} \left(\frac{\mu_E}{I\hbar}\right)^2 \quad (36)$$

where c is the velocity of sound of the crystal.

From the general expression (27) follows (for $l = 2$) the well-known formula for quadrupole rate [13]

$$\begin{aligned} \left(\frac{1}{T_1}\right)_{E2} &= \frac{6}{5} \frac{2I+3}{I^2(2I-1)} c_2^2 (1+a_2) \tau_c = \\ &= \frac{3}{40} \frac{2I+3}{I^2(2I-1)} \left(\frac{e^2 q Q}{\hbar}\right)^2 \left(1 + \frac{\eta^2}{3}\right) \tau_c \end{aligned} \quad (37)$$

where η is the asymmetry parameter related with the factor $a_2 = \eta^2/3$.

Furthermore, for $l = 3$ and 4 one gets from Eq. (27) the following expressions:

$$\left(\frac{1}{T_1}\right)_{M3} = \frac{12}{7} \frac{(2I+3)(I+2)}{I^2(2I-1)(I-1)} c_3^2 (1+a_3) \tau_c \quad (38)$$

$$c_3 = \frac{1}{6} \Omega \frac{\partial^2 H_z}{\partial z^2} \quad (39)$$

$$\left(\frac{1}{T_1}\right)_{E4} = \frac{20}{9} \frac{(2I+3)(I+2)(2I+5)}{I^2(2I-1)(I-1)(2I-3)} c_4^2 (1+a_4) \tau_c \quad (40)$$

$$c_4 = \frac{1}{24} Q^{(4)} \frac{\partial^4 V}{\partial z^4} \quad (41)$$

The expressions (38)–(41) can be important from the theoretical point of view. In practice, the contributions to the relaxation rate produced by magnetic octupole ($M3$) and electric hexadecapole ($E4$) interactions are usually negligible compared with the contributions following from $M1$ and $E2$ interactions, because the coupling constants c_3 and c_4 are very small. From a very rough evaluation [1] it follows that $c_3/c_1 \sim c_4/c_2 \sim R^2/a^2 \sim 10^{-5}$, where R is the nuclear radius and $a = a_0/Z$ is the effective radius of the electronic shell.

The presented theory of multipole relaxations ($l = 2, 3, 4, \dots$) is valid only in the extreme narrowing case (rapid motion) for $\omega_0 \tau_c \ll 1$ and may be applied in liquids of low viscosity and (with some restrictions) in dense gases. If molecular motion is not rapid

the multipole relaxations can be nonexponential. In the case of quadrupole interaction it was shown [14] that the relaxation function is the sum of I decaying exponential terms if spin I is a integer or the sum of $I + \frac{1}{2}$ the decaying exponential terms if I is half an odd integer.

Generally, in the presence of arbitrary multipole interactions the relaxation function $R(t)$ following from the Eqs (12)–(23) and (29) may be written in the form

$$R(t) = \sum_k C_k e^{-\lambda_k \frac{t}{T_1}} \quad (42)$$

where

$$\sum_k C_k = \sum_k \lambda_k C_k = 1 \quad (43)$$

$$-\left(\frac{d \ln R(t)}{dt}\right)_{t=0} = \frac{1}{T_1} = \sum_l \frac{3c_l^2(1-a_l)}{2(2l+1)^2 I(I+1)} \times \\ \times \sum_{m=-l}^l m^2 j(m\omega_0) \prod_{n=1}^l \left(\frac{2I+n+1}{2I-n+1}\right). \quad (44)$$

In the case of $\omega_0\tau_c \ll 1$ the Eqs (42) and (44) lead to the same expressions as those presented in the Eqs (26) and (27)–(28) respectively. For greater values of $\omega_0\tau_c$ (e. g. $\omega_0\tau_c \sim 1$) one should observe a deviation of the relaxation function from the single exponential decay. The number of the exponential terms in Eq. (42) increases with increasing number l and I . To find the values λ_k and C_k in Eq. (42) one has to calculate the eigenvalues and eigenvectors of the matrix $R_{\alpha\beta}$. A method of numerical solution of this problem was presented elsewhere [15].

APPENDIX

An arbitrary electric (El) and magnetic (MI) multipole interaction of the order l may be written as a scalar product of two irreducible spherical tensors $A_{lm}(r_m)$ and $F_{lm}(r_e)$ [1]:

$$\hbar G^{(l)} = \sum_{m=-l}^l (-1)^m A_{lm}(r_n) F_{l-m}(r_e). \quad (A1)$$

The expectation values of the above tensor elements may be presented in the form:

$$\langle A_{lm} \rangle = \int_{\tau_n} f_n r_n^l C_{lm}(\theta_n, \Phi_n) d\tau_n \quad (A2)$$

$$\langle F_{lm} \rangle = \int_{\tau_e} f_e r_e^{-(l+1)} C_{lm}(\theta_e, \Phi_e) d\tau_e \quad (A3)$$

$$C_{lm}(\theta, \Phi) = \sqrt{\frac{4\pi}{2l+1}} Y_{lm}(\theta, \Phi) \quad (A4)$$

$$f_s = \begin{cases} \rho_s & (\text{for } El, s = e, n) \\ -\nabla \cdot m_s & (\text{for } MI, s = e, n) \end{cases} \quad (A5)$$

$$(A6)$$

where $Y_{lm}(\theta, \Phi)$ are spherical harmonics, ρ_s is the density of the electric charge and m_s is the density of magnetization for a nucleus (n) and electronic cloud (e), and r_n, r_e are nuclear and electron position coordinates.

To find the effective spin Hamiltonian $G_{\text{eff}}^{(l)}$ which gives the same matrix elements as $G^{(l)}$ one has to introduce the tensor operators $A_{lm}(\mathbf{I})$ in place of the spherical tensors $A_{lm}(\mathbf{r}_n)$. These tensor operators can be constructed by polarization of the spherical tensors $A_{lm}(\mathbf{r}_n)$, [2], which are proportional to the harmonic polynomials $r^l C_{lm}$. Neglecting the constant factor one can express the tensor operators in the form [16]:

$$A_{lm}(\mathbf{I}) = (\mathbf{I} \cdot \nabla)^l r^l C_{lm}. \quad (\text{A7})$$

The constant factor C may be found from the following condition for the matrix elements:

$$C \langle II | A_{l0}(\mathbf{I}) | II \rangle = \langle II | A_{l0}(r_n) | II \rangle \equiv M_n^{(l)}. \quad (\text{A8})$$

Moreover, the time dependent elements of the spherical tensor $F_{lm}(t) = F_{lm}(\mathbf{r}_n(t))$ in the laboratory reference frame can be expressed by the constant elements F_{lm}^0 in the molecular reference frame [2], [17]

$$F_{lm}(t) = \sum_{m'} D_{m'm}^{(l)}(\alpha, \beta, \gamma) F_{lm'}^0. \quad (\text{A9})$$

with

$$D_{0m}^{(l)}(\alpha, \beta, \gamma) = C_{lm}(\beta, \gamma) \quad (\text{A10})$$

where α, β, γ are the time dependent Euler angles.

In the case of axial symmetry one gets:

$$F_{lm}^0 = \delta_{m0} F_{l0}^0 \quad (\text{A11})$$

$$F_{l0}^0 = \frac{1}{l!} \frac{\partial^l V}{\partial z^l} = -\frac{1}{l!} \frac{\partial^{l-1} E_z}{\partial z^{l-1}} \quad (\text{for } El) \quad (\text{A12})$$

$$F_{l0}^0 = -\frac{1}{l!} \frac{\partial^{l-1} H_z}{\partial z^{l-1}} \quad (\text{for } Ml) \quad (\text{A13})$$

where \mathbf{E} and \mathbf{H} are the local electric and magnetic fields respectively and V is the electric potential.

Using the Eqs (A1) and (A7)–(A13) and $\alpha = \theta(t)$, $\gamma = \Phi(t)$ one can express the effective spin Hamiltonian for the interaction $M3$ and $E4$ in the following form:

$$G^{(M3)}(t) = \frac{2c_3}{I(2I-1)(I-1)} \sum_{m=-3}^3 (-1)^m A_{3m}(\mathbf{I}) \dot{C}_{3-m}(\theta(t), \Phi(t)) \quad (\text{A14})$$

$$G^{(E4)}(t) = \frac{4c_4}{I(2I-1)(I-1)(2I-3)} \sum_{m=-4}^4 (-1)^m A_{4m}(I) C_{4-m}(\theta(t), \Phi(t)) \quad (\text{A15})$$

$$A_{30}(I) = \frac{1}{2} \{5I_z^3 - I_z[3I(I+1) - 1]\} \quad (\text{A16})$$

$$A_{31}(I) = -\frac{\sqrt{3}}{4} [5I_z^2 - 5I_z - I(I+1) + 2] I_+ \quad (\text{A17})$$

$$\begin{aligned} A_{32}(I) &= \frac{\sqrt{30}}{12} (I_z I_+^2 + I_+ I_z I_+ + I_+^2 I_z) = \\ &= \frac{\sqrt{30}}{4} (I_z - 1) I_+^2 \end{aligned} \quad (\text{A18})$$

$$A_{33}(I) = -\frac{\sqrt{3}}{4} I_+^3 \quad (\text{A19})$$

$$\begin{aligned} A_{40}(I) &= \frac{1}{8} \{35I_z^4 + I_z^2[30I(I+1) - 25] + \\ &+ 3I(I+1)[I(I+1) - 2]\} \end{aligned} \quad (\text{A20})$$

$$\begin{aligned} A_{41}(I) &= -\frac{\sqrt{5}}{4} [I_z^3 I_+ + I_z^2 I_+ I_z + I_z I_+ I_z^2 + I_+ I_z^3 - \\ &- \frac{1}{4} (I_z I_+ I_+ I_+ + I_z I_+ I_+^2 + I_z I_+^2 I_+ + I_+ I_z I_+ I_+ + \\ &+ I_+ I_+ I_z I_+ + I_+ I_z I_+ I_+ + I_+ I_+ I_+ I_z + I_+^2 I_z I_+ + \\ &+ I_+^2 I_+ I_z + I_+ I_z I_+^2 + I_+ I_+ I_z I_+ + I_+ I_+^2 I_z)] \end{aligned} \quad (\text{A21})$$

$$\begin{aligned} A_{42}(I) &= \frac{\sqrt{10}}{4} [I_z^2 I_+^2 + I_z I_+ I_z I_+ + I_z I_+^2 I_z + \\ &+ I_+ I_z^2 I_+ + I_+ I_z I_+ I_z + I_+^2 I_z^2 - \\ &- \frac{1}{4} (I_+^3 I_+ + I_+^2 I_+ I_+ + I_+ I_+ I_+^2 + I_+ I_+^3)] \end{aligned} \quad (\text{A22})$$

$$A_{43}(I) = -\frac{\sqrt{35}}{16} (I_+^3 I_z + I_+^2 I_z I_+ + I_+ I_z I_+^2 + I_z I_+^3) \quad (\text{A23})$$

$$A_{44}(I) = \frac{\sqrt{70}}{16} I_+^4 \quad (\text{A24})$$

$$A_{l-m}(I) = (-1)^m A_{lm}^*(I). \quad (\text{A25})$$

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