

SUPPLEMENTARY NOTE TO THE PAPER:  
MOLECULAR THEORY OF LIGHT SCATTERING BY MULTI-  
-COMPONENT SYSTEMS

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The present note is to supplement the paper on a "Molecular Theory of Light Scattering by Multi-Component Systems" (Kielich, 1960), and brings a discussion of the scattered light intensity for the case of a system whose various components consist of quadrupolar, anisotropically polarizable and hyperpolarizable molecules.

The light intensity scattered at the angle  $\vartheta$  is given by the general formula (see, Kielich, 1960)

$$I(\vartheta) = \frac{8\pi^4 I_0}{45 \lambda^4 R_0^2} \{5(1 + \cos^2 \vartheta) F_{\text{is}}(s) + (13 + \cos^2 \vartheta) F_{\text{anis}}(s)\} \quad (1)$$

where  $I_0$  and  $\lambda$  denote the intensity and wave-length of the incident light, respectively and  $R_0$  is the distance of the point of observation from the centre of the scattering volume  $V$ . The factors  $F_{\text{is}}(s)$  and  $F_{\text{anis}}(s)$  characterize the molecular mechanism of isotropic and anisotropic light scattering, respectively. For a multi-component system these are of the form

$$F_{\text{is}}(s) = \sum_{i,j} \left\langle \delta_{\alpha\beta} \delta_{\gamma\delta} \sum_{p=1}^{x_i N} \sum_{q=1}^{x_j N} \frac{\partial m_{\alpha}^{(p,i)}}{\partial E_{\beta}^{(p,i)}} \left( \frac{\partial m_{\gamma}^{(q,j)}}{\partial E_{\delta}^{(q,j)}} \right)^* e^{-is \cdot \mathbf{r}_{ij}^{(pq)}} \right\rangle, \quad (2)$$

$$F_{\text{anis}}(s) = \frac{1}{2} \sum_{i,j} \left\langle (3\delta_{\alpha\gamma} \delta_{\beta\delta} - \delta_{\alpha\beta} \delta_{\gamma\delta}) \sum_{p=1}^{x_i N} \sum_{q=1}^{x_j N} \frac{\partial m_{\alpha}^{(p,i)}}{\partial E_{\beta}^{(p,i)}} \left( \frac{\partial m_{\gamma}^{(q,j)}}{\partial E_{\delta}^{(q,j)}} \right)^* e^{-is \cdot \mathbf{r}_{ij}^{(pq)}} \right\rangle, \quad (3)$$

where  $x_i$  is the mole fraction of the  $i$ -th component,  $m_{\alpha}^{(p,i)}$  — the  $\alpha$ -component of the electric dipole moment induced by the electric field  $E_{\alpha}$  of the incident light wave in the  $p$ -th molecule of species  $i$ ,  $\mathbf{r}_{ij}^{(pq)}$  — the vector connecting the centres of the

$p$ -th and  $q$ -th molecules of the  $i$ -th and  $j$ -th species, and, for Rayleigh scattering,  $|\mathbf{s}| = (4\pi/\lambda) \sin(\vartheta/2)$ . The brackets  $\langle \quad \rangle$  in eqs. (2) and (3) denote the statistical average in the absence of the external electric field ( $\mathbf{E} = 0$ ) and  $\delta_{\alpha\beta}$  is the substitution tensor (unity if  $\alpha = \beta$  and zero when  $\alpha \neq \beta$ ).

The total differential polarizability tensor of the molecule immersed in the medium is given by the following expansion (for  $\mathbf{E} = 0$ ):

$$\left( \frac{\partial m_{\alpha}^{(p,i)}}{\partial E_{\chi}^{(p,i)}} \right)_{\mathbf{E}=0} = \left\{ \alpha_{\alpha\beta}^{(p,i)} + \beta_{\alpha\beta\gamma}^{(p,i)} F_{\gamma}^{(p,i)} + \frac{1}{2} \gamma_{\alpha\beta\gamma\delta}^{(p,i)} F_{\gamma}^{(p,i)} F_{\delta}^{(p,i)} + \frac{1}{3} B_{\alpha\beta:\gamma\delta}^{(p,i)} F_{\gamma\delta}^{(p,i)} + \dots \right\} \left\{ \delta_{\beta\chi} + \frac{\partial F_{\beta}^{(p,i)}}{\partial E_{\chi}^{(p,i)}} \right\}_{\mathbf{E}=0}, \quad (4)$$

with  $\alpha_{\alpha\beta}^{(p,i)}$  denoting the polarizability tensor of the  $p$ -th molecule of species  $i$ , and  $\beta_{\alpha\beta\gamma}^{(p,i)}$ ,  $\gamma_{\alpha\beta\gamma\delta}^{(p,i)}$  —its hyperpolarizability tensors. The tensor  $B_{\alpha\beta:\gamma\delta}^{(p,i)}$  accounts for the additional polarization, of the  $p$ -th molecule of species  $i$  as induced by the field gradient  $F_{\alpha\beta}^{(p,i)}$ . The tensors  $\alpha_{\alpha\beta}$ ,  $\beta_{\alpha\beta\gamma}$ ,  $\gamma_{\alpha\beta\gamma\delta}$  and  $B_{\alpha\beta:\gamma\delta}$  have been discussed by Buckingham (1959).  $F_{\alpha}^{(p,i)}$  and  $F_{\alpha\beta}^{(p,i)}$  are the  $\alpha$ -component of the molecular field and  $\alpha\beta$ -component of the field gradient at the centre of molecule  $p$  of species  $i$  due to the electric charge distributions of all the others, in the presence of the electric field  $E_{\alpha}$ .

Substituting the expansion of eq. (4) in eqs. (2) and (3), the following expressions are obtained:

$$F_{\text{is}}(s) = \sum_{i,j} \left\langle \sum_{p=1}^{x_i N} \sum_{q=1}^{x_j N} \{ \alpha_{\alpha\alpha}^{(i)} \alpha_{\beta\beta}^{(j)} + A_{\alpha\alpha:\beta\beta}^{(ij)} + H_{\alpha\alpha:\beta\beta}^{(ij)} \} \cos \mathbf{s} \cdot \mathbf{r}_{ij}^{(pq)} \right\rangle \quad (5)$$

$$F_{\text{anis}}(s) = \frac{1}{2} \sum_{i,j} \left\langle \sum_{p=1}^{x_i N} \sum_{q=1}^{x_j N} \{ \alpha_{\alpha\beta}^{(i)} \alpha_{\gamma\delta}^{(j)} + A_{\alpha\beta:\gamma\delta}^{(ij)} + H_{\alpha\beta:\gamma\delta}^{(ij)} \} \{ 3\omega_{\alpha\gamma}^{(pq;ij)} \omega_{\beta\delta}^{(pq;ij)} - \delta_{\alpha\beta} \delta_{\gamma\delta} \} \cos \mathbf{s} \cdot \mathbf{r}_{ij}^{(pq)} \right\rangle. \quad (6)$$

Here,  $\omega_{\alpha\gamma}^{(pq;ij)}$  denotes the cosine of the angle subtended by the  $\alpha$  and  $\gamma$  axes of the molecular systems of reference rigidly attached to the  $p$ -th and  $q$ -th molecules of species  $i$  and  $j$  respectively. The tensors

$$A_{\alpha\beta:\gamma\delta}^{(ij)} = \alpha_{\alpha\beta}^{(i)} \alpha_{\gamma\delta}^{(j)} \frac{\partial F_{\epsilon}^{(j)}}{\partial E_{\delta}^{(j)}} + \alpha_{\gamma\delta}^{(j)} \alpha_{\alpha\epsilon}^{(i)} \frac{\partial F_{\epsilon}^{(i)}}{\partial E_{\beta}^{(i)}} + \alpha_{\alpha\epsilon}^{(i)} \alpha_{\gamma\eta}^{(j)} \frac{\partial F_{\epsilon}^{(i)}}{\partial E_{\beta}^{(i)}} \frac{\partial F_{\eta}^{(j)}}{\partial E_{\delta}^{(j)}}, \quad (7)$$

$$H_{\alpha\beta:\gamma\delta}^{(ij)} = \alpha_{\alpha\beta}^{(i)} \beta_{\gamma\delta\epsilon}^{(j)} F_{\epsilon}^{(j)} + \alpha_{\gamma\delta}^{(j)} \beta_{\alpha\beta\epsilon}^{(i)} F_{\epsilon}^{(i)} + \dots + \frac{1}{2} \alpha_{\alpha\beta}^{(i)} \gamma_{\gamma\delta\epsilon\eta}^{(j)} F_{\epsilon}^{(j)} F_{\eta}^{(j)} + \frac{1}{2} \alpha_{\gamma\delta}^{(j)} \gamma_{\alpha\beta\epsilon\eta}^{(i)} F_{\epsilon}^{(i)} F_{\eta}^{(i)} + \dots + \frac{1}{3} \alpha_{\alpha\beta}^{(i)} B_{\gamma\delta:\epsilon\eta}^{(j)} F_{\epsilon\eta}^{(j)} + \frac{1}{3} \alpha_{\gamma\delta}^{(j)} B_{\alpha\beta:\epsilon\eta}^{(i)} F_{\epsilon\eta}^{(i)} + \dots \quad (8)$$

determine the effect of the anisotropy of the molecular field and that of the hyperpolarizability of the molecules on light scattering in a dense medium.

With respect to Onsager's (1936) model, the anisotropy of the molecular field is given by

$$\frac{\partial F_{\alpha}^{(i)}}{\partial E_{\beta}^{(i)}} = \psi_{\alpha}^{(i)} \delta_{\alpha\beta} \quad (9)$$

and the molecular field  $F_{\alpha}^{(i)}$  and field gradient  $F_{\alpha\beta}^{(i)}$  at  $\mathbf{E} = 0$  are

$$F_{\alpha}^{(i)} = \chi_{\alpha}^{(i)} \frac{\mu_{\alpha}^{(i)}}{\alpha_{(i)}} + \xi_{\alpha}^{(i)} \frac{\Theta_{\alpha\beta}^{(i)} r_{\beta}}{\alpha_{(i)}^{3/2}}, \quad (10)$$

$$F_{\alpha\beta}^{(i)} = \frac{\partial F_{\alpha}^{(i)}}{\partial r_{\beta}} = \xi_{\alpha}^{(i)} \frac{\Theta_{\alpha\beta}^{(i)}}{\alpha_{(i)}^{3/2}}, \quad (11)$$

wherein  $\mu_{\alpha}^{(i)}$  is the  $\alpha$ -component of the permanent electric dipole moment of an isolated polar molecule of species  $i$  and  $\Theta_{\alpha\beta}^{(i)}$ —its electric quadrupole moment. The parameters  $\psi_{\alpha}^{(i)}$ ,  $\chi_{\alpha}^{(i)}$  and  $\xi_{\alpha}^{(i)}$  are given by:

$$\psi_{\alpha}^{(i)} = \frac{2(n^2 - 1) \{(n^2 + 2)(n_i^2 - 1)\lambda_{\alpha}^{(i)} - (n_i^2 + 2)(n^2 - 1)\}}{(n^2 + 2) \{(2n^2 + 1)(n_i^2 + 2) - 2(n^2 - 1)(n_i^2 - 1)\lambda_{\alpha}^{(i)}\}}, \quad (12)$$

$$\chi_{\alpha}^{(i)} = \frac{2(\varepsilon - 1)(n_i^2 - 1)}{(2\varepsilon + 1)(n_i^2 + 2) - 2(\varepsilon - 1)(n_i^2 - 1)\lambda_{\alpha}^{(i)}}, \quad (13)$$

$$\xi_{\alpha}^{(i)} = 3 \left( \frac{2\varepsilon + 1}{3\varepsilon + 2} \right) \left( \frac{n_i^2 - 1}{n_i^2 + 2} \right)^{3/2} \chi_{\alpha}^{(i)}, \quad (14)$$

where  $\varepsilon$  and  $n$  are the electric permittivity and refractive index of the medium, respectively, and  $\lambda_{\alpha}^{(i)} = \alpha_{\alpha}^{(i)}/\alpha_{(i)}$ , with  $\alpha_{\alpha}^{(i)}$  denoting the polarizability component in the direction of an  $\alpha$ -principal axis of an isolated molecule of species  $i$ , and  $\alpha_{(i)}$ —its mean polarizability.

For non-dipolar molecules ( $\mu_{\alpha} = \beta_{\alpha\beta\gamma} = 0$ ) possessing the axial symmetry, the following tensor elements only are non-vanishing:  $\alpha_{\alpha\beta}$ ,  $\Theta_{\alpha\beta}$ ,  $\gamma_{\alpha\beta\gamma\delta}$  and  $B_{\alpha\beta:\gamma\delta}$  (the axis of symmetry being parallel to 03, see Buckingham 1959):

$$\begin{aligned} \alpha_{11} = \alpha_{22} = \alpha_{\perp}, \quad \gamma_{1111} = \gamma_{2222} = 3\gamma_{1122} = \gamma_{\perp}, \quad \gamma_{1133} = \gamma_{2233} = \gamma_{\parallel}, \\ \alpha_{33} = \alpha_{\parallel}, \quad \gamma_{3333} = \gamma_{\parallel}, \quad \Theta_{33} = -2\Theta_{11} = -2\Theta_{22} = \Theta, \\ B_{33:33} = -2B_{33:11} = -2B_{33:22} = 2(B_{11:11} + B_{11:22}) = B, \end{aligned} \quad (15)$$

where  $\Theta$  is the quadrupole moment of the axially symmetric molecule and  $B$ —its quadrupole polarizability.

With respect to (15) and for  $\lambda \gg r_{ij}$ , the molecular factors of isotropic and anisotropic light scattering assume the form:

$$\begin{aligned} F_{\text{is}} = 9N \sum_{i,j} \alpha_{(i)} \alpha_{(j)} \{1 + A_{\text{is}}^{(ij)} + H_{\text{is}}^{(ij)}\} \times \\ \times \left\{ x_i \delta_{ij} + x_i x_j \int \left[ g_{ij}(\tau) - \frac{N}{V} \right] d\bar{\tau} \right\}, \end{aligned} \quad (16)$$

$$\begin{aligned} F_{\text{anis}} = 9N \sum_{i,j} \alpha_{(i)} \delta_{\alpha(i)} \alpha_{(j)} \delta_{\alpha(j)} \{1 + A_{\text{anis}}^{(ij)} + H_{\text{anis}}^{(ij)}\} \times \\ \times \left\{ x_i \delta_{ij} + \frac{1}{2} x_i x_j \int (3\cos^2 \Theta_{ij} - 1) \left[ g_{ij}(\tau) - \frac{N}{V} \right] d\bar{\tau} \right\}, \end{aligned} \quad (17)$$

which holds for quadrupole molecules having the axial symmetry. Here,  $\Theta_{ij}$  is the angle between the axes of symmetry of the molecules of species  $i$  and  $j$ , respectively,  $g_{ij}(\tau)$  – the distribution function and  $\Omega^2 d\tau = dr_{ij} d\omega_i d\omega_j$ , where  $\Omega = \int d\omega_i$ . The quantities  $A_{\text{is}}^{(ij)}$  and  $A_{\text{anis}}^{(ij)}$  in the foregoing expressions account for the effect of the anisotropy of the molecular field on isotropic and anisotropic light scattering, respectively, and are given by

$$A_{\text{is}}^{(ij)} = \frac{1}{8} (\lambda_{\parallel}^{(i)} \psi_{\parallel}^{(i)} + 2\lambda_{\perp}^{(i)} \psi_{\perp}^{(i)}) + \frac{1}{8} (\lambda_{\parallel}^{(j)} \psi_{\parallel}^{(j)} + 2\lambda_{\perp}^{(j)} \psi_{\perp}^{(j)}) + \\ + \frac{1}{8} (\lambda_{\parallel}^{(i)} \psi_{\parallel}^{(i)} + 2\lambda_{\perp}^{(i)} \psi_{\perp}^{(i)}) (\lambda_{\parallel}^{(j)} \psi_{\parallel}^{(j)} + 2\lambda_{\perp}^{(j)} \psi_{\perp}^{(j)}), \quad (18)$$

$$A_{\text{anis}}^{(ij)} = \frac{\lambda_{\parallel}^{(i)} \psi_{\parallel}^{(i)} - \lambda_{\perp}^{(i)} \psi_{\perp}^{(i)}}{\lambda_{\parallel}^{(i)} - \lambda_{\perp}^{(i)}} + \frac{\lambda_{\parallel}^{(j)} \psi_{\parallel}^{(j)} - \lambda_{\perp}^{(j)} \psi_{\perp}^{(j)}}{\lambda_{\parallel}^{(j)} - \lambda_{\perp}^{(j)}} + \\ + \left( \frac{\lambda_{\parallel}^{(i)} \psi_{\parallel}^{(i)} - \lambda_{\perp}^{(i)} \psi_{\perp}^{(i)}}{\lambda_{\parallel}^{(i)} - \lambda_{\perp}^{(i)}} \right) \left( \frac{\lambda_{\parallel}^{(j)} \psi_{\parallel}^{(j)} - \lambda_{\perp}^{(j)} \psi_{\perp}^{(j)}}{\lambda_{\parallel}^{(j)} - \lambda_{\perp}^{(j)}} \right). \quad (19)$$

$H_{\text{is}}^{(ij)}$  and  $H_{\text{anis}}^{(ij)}$  account for the additional contribution to isotropic and anisotropic light scattering due to higher order effects such as hyperpolarizability of the molecules and polarizability of molecular quadrupoles. For these quantities we have:

$$H_{\text{is}}^{(ij)} = \frac{5}{2} \eta_{(i)} \kappa_{\text{is}}^{(i)} \frac{\gamma_{(i)} \Theta_{(i)}^2}{\alpha_{(i)}^{11/2}} + \frac{5}{2} \eta_{(j)} \kappa_{\text{is}}^{(j)} \frac{\gamma_{(j)} \Theta_{(j)}^2}{\alpha_{(j)}^{11/2}} + \dots, \quad (20)$$

$$H_{\text{anis}}^{(ij)} = \frac{7}{4} \eta_{(i)} \kappa_{\text{anis}}^{(i)} \frac{\gamma_{(i)} \delta_{\gamma(i)} \Theta_{(i)}^2}{\alpha_{(i)}^{11/2} \delta_{\alpha(i)}} + \frac{7}{4} \eta_{(j)} \kappa_{\text{anis}}^{(j)} \frac{\gamma_{(j)} \delta_{\gamma(j)} \Theta_{(j)}^2}{\alpha_{(j)}^{11/2} \delta_{\alpha(j)}} + \\ + \frac{1}{4} \xi_{(i)} \frac{B_{(i)} \Theta_{(i)}}{\alpha_{(i)}^{9/2} \delta_{\alpha(i)}} + \frac{1}{4} \xi_{(j)} \frac{B_{(j)} \Theta_{(j)}}{\alpha_{(j)}^{9/2} \delta_{\alpha(j)}} + \dots \quad (21)$$

Here

$$\delta_{\alpha(i)} = \frac{\alpha_{\parallel}^{(i)} - \alpha_{\perp}^{(i)}}{3\alpha_{(i)}} \quad \text{and} \quad \alpha_{(i)} = \frac{1}{3} (\alpha_{\parallel}^{(i)} + 2\alpha_{\perp}^{(i)}) \quad (22)$$

describe the anisotropy of polarizability of the isolated molecule of species “ $i$ ” and its mean polarizability, respectively, whereas

$$\delta_{\gamma(i)} = \frac{2(3\gamma_{\parallel}^{(i)} + 3\gamma_{\perp}^{(i)} - 4\gamma_{\perp}^{(i)})}{21\gamma_{(i)}}, \quad \gamma_{(i)} = \frac{1}{15} (3\gamma_{\parallel}^{(i)} + 12\gamma_{\perp}^{(i)} + 8\gamma_{\perp}^{(i)}) \quad (23)$$

are the anisotropy of hyperpolarizability of the molecule of species  $i$  and its mean hyperpolarizability.

The parameters  $\eta_{(i)}$ ,  $\xi_{(i)}$ ,  $\kappa_{\text{is}}^{(i)}$  and  $\kappa_{\text{anis}}^{(i)}$ , introduced in eqs. (20) and (21) are defined as follows

$$\eta_{(i)} = \frac{1}{10} \left( \frac{2\varepsilon + 1}{3\varepsilon + 2} \right)^2 \left( \frac{n_i^2 - 1}{n_i^2 + 2} \right)^{3/2} (\chi_{\perp}^{(i)^2} + 2\chi_{\parallel}^{(i)^2}), \quad (24)$$

$$\xi^{(i)} = \left( \frac{2\varepsilon + 1}{3\varepsilon + 2} \right) \left( \frac{n_i^2 - 1}{n_i^2 + 2} \right)^{3/2} (\chi_{\perp}^{(i)} + 2\chi_{\parallel}^{(i)}), \quad (25)$$

$$\kappa_{\text{is}}^{(i)} = \{15\gamma_{(i)} \overline{F_{i3}^2} - 2(3\gamma_{\parallel}^{(i)} + 4\gamma_{\perp}^{(i)}) (\overline{F_{i3}^2} - \overline{F_{i1}^2})\} \{5\gamma_{(i)} \overline{F_i^2}\}^{-1}, \quad (26)$$

$$\kappa_{\text{anis}}^{(i)} = \{21\gamma_{(i)} \delta_{\gamma(i)} \overline{F_{i3}^2} - 4(3\gamma_{\parallel}^{(i)} - 2\gamma_{\perp}^{(i)}) (\overline{F_{i3}^2} - \overline{F_{i1}^2})\} \{7\gamma_{(i)} \delta_{\gamma(i)} \overline{F_i^2}\}^{-1}, \quad (27)$$

wherein the mean square molecular fields  $\overline{F_{i1}^2}$ ,  $\overline{F_{i3}^2}$  and  $\overline{F_i^2}$  are given, for axially symmetric quadrupolar molecules, by:

$$\overline{F_{i1}^2} = \overline{F_{i2}^2} = \frac{9}{20} \left( \frac{2\varepsilon + 1}{3\varepsilon + 2} \right)^2 \left( \frac{n_i^2 - 1}{n_i^2 + 2} \right)^{3/2} \chi_{\perp}^{(i)} \frac{\Theta_{(i)}^2}{\alpha_{(i)}^{3/2}}, \quad (28)$$

$$\overline{F_{i3}^2} = \frac{9}{5} \left( \frac{2\varepsilon + 1}{3\varepsilon + 2} \right)^2 \left( \frac{n_i^2 - 1}{n_i^2 + 2} \right)^{3/2} \chi_{\parallel}^{(i)} \frac{\Theta_{(i)}^2}{\alpha_{(i)}^{3/2}}, \quad (29)$$

$$\overline{F_i^2} = 2\overline{F_{i1}^2} + \overline{F_{i3}^2} = 9\eta_{(i)} \frac{\Theta_{(i)}^2}{\alpha_{(i)}^{3/2}}. \quad (30)$$

In a former paper (Kielich 1960), when passing from the formulas (5.2) and (5.3) to eqs. (5.13) and (5.14), the simplifying assumption of  $\kappa_{\text{is}}^{(i)} = \kappa_{\text{anis}}^{(i)} = 1$  had been made. From the foregoing eqs. (26) and (27), this condition is seen to be satisfied strictly only if the molecular field possesses the spherical symmetry, i. e. if  $\overline{F_{i1}^2} = \overline{F_{i2}^2} = \overline{F_{i3}^2} = \frac{1}{3} \overline{F_i^2}$ . Assuming also  $\gamma_{\parallel}^{(i)} = \frac{1}{3} (\gamma_{\parallel}^{(i)} + 2\gamma_{\perp}^{(i)})$  (see, Buckingham and Stephen 1957), the quantities  $\delta_{\gamma(i)}$  and  $\gamma_{(i)}$  defined above by eqs. (23) reduce to

$$\delta_{\gamma(i)} = \frac{\gamma_{\parallel}^{(i)} - \gamma_{\perp}^{(i)}}{3\gamma_{(i)}}, \quad \gamma_{(i)} = \frac{1}{3} (\gamma_{\parallel}^{(i)} + 2\gamma_{\perp}^{(i)}) \quad (31)$$

which is identical with the expressions of (5.15) (Kielich, 1960).

Also, eq. (7.5) for the molecular refraction  $R_m$  was derived on the simplifying assumption of  $\kappa_{\text{is}}^{(i)} = 1$ . Without the latter, we have the general expression

$$R_m = \frac{4\pi}{3} N \sum_i x_i \alpha_{(i)} \left\{ 1 + \frac{1}{3} (\lambda_{\parallel}^{(i)} \psi_{\parallel}^{(i)} + 2\lambda_{\perp}^{(i)} \psi_{\perp}^{(i)}) + \frac{5}{2} \eta_{(i)} \kappa_{\text{is}}^{(i)} \frac{\gamma_{(i)} \Theta_{(i)}^2}{\alpha_{(i)}^{3/2}} \right\}, \quad (32)$$

which holds for axially symmetric quadrupolar molecules.

In the case of a one-component system, eqs. (16), (17) and (32) reduce to the form

$$F_{\text{is}} = 9\alpha^2 N \left\{ \left( 1 + \frac{\lambda_{\parallel} \psi_{\parallel} + 2\lambda_{\perp} \psi_{\perp}}{3} \right)^2 + 5\eta \kappa_{\text{is}} \frac{\gamma \Theta^2}{\alpha^{3/2}} \right\} \frac{RT\beta_T}{V}, \quad (33)$$

$$F_{\text{anis}} = 9\alpha^2 \delta_{\alpha}^2 N \left\{ \left( 1 + \frac{\lambda_{\parallel} \psi_{\parallel} - \lambda_{\perp} \psi_{\perp}}{\lambda_{\parallel} - \lambda_{\perp}} \right)^2 + \frac{7}{2} \eta \kappa_{\text{anis}} \frac{\gamma \delta_{\gamma} \Theta^2}{\alpha^{3/2} \delta_{\alpha}} + \frac{1}{2} \xi \frac{B\Theta}{\alpha^{3/2} \delta_{\alpha}} \right\} \left\{ 1 + \frac{1}{2} \int (3 \cos^2 \Theta - 1) \left[ g(\tau) - \frac{N}{V} \right] d\tau \right\}, \quad (34)$$

$$R_m = \frac{4\pi}{3} N\alpha \left\{ 1 + \frac{1}{3} (\lambda_{\parallel} \psi_{\parallel} + 2\lambda_{\perp} \psi_{\perp}) + \frac{5}{2} \eta_{\text{is}} \frac{\gamma \Theta^2}{\alpha^{11/3}} \right\}, \quad (35)$$

with  $R$  denoting the gas constant,  $T$ —the absolute temperature,  $\beta_T$ —the isothermal compressibility coefficient of the medium and  $V$ —its molar volume.

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