

ON THE IMPACT APPROXIMATION THEORY OF SPECTRUM SHAPE RELAXATION PARAMETERS

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Received December 23, 1993

This paper presents a modification of the method earlier proposed by the author for calculating the set of relaxation parameters describing, in the impact approximation, the shape of a spectrum. The modification proposed in this paper makes the method easier for computations and widens its applicability limits. To illustrate the calculational efficiency of the modification, the data of calculations of self-broadening for some rotational doublets of ammonia are presented.

1. INTRODUCTION

Collisional, or pressure, line broadening is the most essential mechanism for forming spectral line profiles, as well as the whole spectrum in a wide range of thermodynamic conditions. A central part of spectral lines is usually described using the impact approximation of the theory, which is valid for not too dense gaseous media. In this approximation the shape of a spectrum is characterized by set of frequency-independent parameters that are the reduced matrix elements of the relaxation superoperator¹⁾ acting in the spectral line space representation.¹ In approximation of classical trajectories the superoperator has a form

$$\hat{\Lambda} = -i \eta_b \int dv P(v) \text{Tr}^b \{ [1 - \hat{U}(\infty, -\infty)] \rho^b \}, \quad (1)$$

where η_b is the buffer gas density, this gas acts as a thermostat for radiation absorbing molecules, $\int dv P(v)$ is the averaging operator over classical collision of parameters, the trace Tr^b is taken over all states of the thermostat particle excluding the translation ones, ρ^b is the density matrix of the particle, and $\hat{U}(\infty, -\infty)$ is the scattering superoperator in Liouville space of the light-absorbing molecule and the thermostat particle, which is the solution of the evolution equation^{2,3}

$$\frac{\partial \hat{U}(t, t_0)}{\partial t} = -i \hat{L}_c(t) \hat{U}(t, t_0), \quad \hat{U}(t_0, t_0) = \hat{1}, \quad (2)$$

at $t \rightarrow +\infty$ and $t_0 \rightarrow -\infty$. Here $\hat{L}_c(t)$ is the Liouville interaction superoperator, or Liouvillian, written in the interaction representation. In the case of $2K$ -pole radiation of π -parity (for the electric dipole radiation the indices are $K = 1$ and $\pi = -1$) the reduced matrix element

1) The superoperators are defined in the Hilbert space generated by ordinary operators which act in a Hilbert space of wave vectors, with the scalar product $(A, B) = \text{Tr} \{ A^\dagger B \}$. Below they are marked by caret $\hat{}$.

of the relaxation superoperator $\hat{\Lambda}$ is defined by the formula given in Refs. 16 and 2²⁾

$$\hat{\Lambda}_{i'f, if}^{(\pi K)} = \sum_{(m_i m_f)} \frac{(j_f K m_f Q | j_i m_i) (j_f K m_f Q | j_i m_i)}{[(2j_i + 1)(2j_i + 1)]^{1/2}}, \quad (3)$$

$$\ll \alpha_{i' j_i m_i} (\alpha_{i' j_f m_f})^+ | \hat{\Lambda} | \alpha_i j_i m_i (\alpha_f j_f m_f)^+ \gg.$$

Here the subscripts i, i' and f, f' mark the initial final states of the transitions, respectively; j is the quantum number of the total angular momentum, m is its projection on an axis fixed in space; α is a concise designation for all the rest quantum numbers; the quantities of $(\alpha \beta \gamma \delta | \epsilon \phi)$ type are the Clebsch-Gordon coefficients. Enclosing the summing indices in Eq. (3) in parentheses means that the summation is performed over all enclosed indices, both marked and unmarked by a prime.

The diagonal matrix elements of the superoperator $\hat{\Lambda}$ determine the halfwidths and the shifts of separate lines in a spectrum. The nondiagonal matrix elements reflect the collision-induced correlation among the lines (i.e., the collisional interference), and they are responsible for a number of anomalies observed in spectral line profiles, especially when the latter are transformed by pressure.⁴⁻⁷

According to Eq. (1), calculations of the impact relaxation parameters are connected closely with calculations of the matrix elements of the scattering superoperator $\hat{U}(\infty, -\infty)$. Naturally, the latter can be calculated using any known methods of calculation of scattering matrices represented in a superoperator formalism. The most direct way for these calculations is to use the iteration series of a perturbation theory. It is precisely this method of calculation of impact relaxation parameters which had been developed in Ref. 8. In a special case of isolated spectral lines it is reduced to the known Anderson-Tsao-Curnutte-Frost method of calculation of the line halfwidths and shifts of line centres.⁹⁻¹¹ Nevertheless, this way faces some problems associated with

2) For vectors in the spectral line space we use the designation $|if+\rangle = |i\rangle\langle f|$ borrowed from Ref. 1.

the divergence of the integral over the aiming parameter on its lower limit, the elimination of which requires the application of poorly conditioned artificial methods.^{9,10} Due to this reason, the methods leading to an exponential representation of the matrix element are preferable, and that representation can be considered as a partial summation of the iteration series. Similar methods were used, for example, in Refs. 3, 12–15. We develop below the version used in Ref. 15.

2. REDUCED MATRIX ELEMENTS OF THE IMPACT RELAXATION OPERATOR

Our immediate task is to represent the reduced matrix element of $\hat{\Lambda}$ operator in terms of the reduced matrix elements of the scattering superoperator $\hat{U}(\infty, -\infty)$. Bearing it in mind, let us determine the vector in the Liouville space of a light-absorbing molecule as follows:

$$\begin{aligned} & |\alpha_i j_i (\alpha_f j_f)^+; \pi K Q \gg = \\ & = \sum_{m_i m_f} \left(\frac{2K+1}{2j_i+1} \right)^{1/2} (j_f K m_f Q | j_i m_i) |\alpha_i j_i m_i (\alpha_f j_f m_f)^+ \gg, \quad (4) \end{aligned}$$

where $\pi = \pi_i \times \pi_f$ (the parity indices enter into the set of quantum numbers α_i and α_f). Formula (3) can be written now as

$$\begin{aligned} & \hat{\Lambda}_{i'f'}^{(\pi K)} = \frac{1}{2K+1} \sum \ll \alpha_{i'} j_{i'} (\alpha_f j_f)^+; \\ & \pi K Q | \hat{\Lambda} | \alpha_i j_i (\alpha_f j_f)^+; \pi K Q \gg. \quad (5) \end{aligned}$$

Note, that the vectors in Eq. (4) correspond formally to the momentum coupling scheme $\mathbf{j}_f - \mathbf{j}_i = \mathbf{K}$, and they transform according to the irreducible representation (πK) of the rotation–inversion group.

Rewrite then the trace from Eq. (1) following Ref. 16

$$\text{Tr}^b \{ [1 - \hat{U}(\infty, -\infty)] \rho^b \} = \ll 1^b | 1 - \hat{U}(\infty, -\infty) | \rho^b \gg, \quad (6)$$

where the following vectors are introduced:

$$|1^b \gg = \sum_{\beta l} (2l+1)^{1/2} | \beta l (\beta l)^+; 00 \gg, \quad (7)$$

$$|\rho^b \gg = \sum_{\beta l} (2l+1)^{-1/2} \rho_{\beta l}^b | \beta l (\beta l)^+; 00 \gg, \quad (8)$$

and the populations $\rho_{\beta l}^b$ are defined by the formula

$$\rho_{\beta l}^b = \frac{S_{\beta l} (2l+1) \exp \{-E_{\beta l} / (k_B T)\}}{\sum_{\beta l} S_{\beta l} (2l+1) \exp \{-E_{\beta l} / (k_B T)\}}. \quad (9)$$

Here k_B is the Boltzman constant, T is the gas temperature, and $E_{\beta l}$ are the energies of the levels βl .

The orthonormal vectors $|\beta l (\beta l)^+; 00 \gg$ are defined by the formula

$$|\beta l (\beta l)^+; 00 \gg = (2l+1)^{-1/2} \sum |\beta l \mu (\beta l \mu)^+; 00 \gg \quad (10)$$

and correspond to the momentum coupling scheme $\mathbf{l}_f - \mathbf{l}_i = 0$. The latter is a direct consequence of their use only in the operation of averaging over thermostat particle states.

Using Eqs. (6)–(8) we obtain the reduced matrix element of the impact relaxation operator

$$\begin{aligned} & \hat{\Lambda}_{nm}^{(\pi K)} = -i \eta_b \int dv P(v) \left\{ \delta_{nm} - \sum_{\beta l} \rho_{\beta l}^b \left(\frac{2l+1}{2l+1} \right)^{1/2} \times \right. \\ & \left. \times \ll \hat{n}(\hat{\gamma}) | \hat{U}^{(\pi K)}(\infty, -\infty) | (\hat{\beta}) \hat{m} \gg \right\}, \quad (11) \end{aligned}$$

where for brevity sake, the following definitions are made

$$\begin{aligned} & |\hat{m} \gg = |\alpha_i j_i (\alpha_f j_f)^+; \pi K Q \gg, \\ & |\hat{n} \gg = |\alpha_{i'} j_{i'} (\alpha_f j_f)^+; \pi K Q \gg, \\ & |\hat{\beta} \gg = |\beta l (\beta l)^+; 00 \gg, \quad |\hat{\gamma} \gg = |\beta l (\beta l)^+; 00 \gg, \\ & |(\hat{\beta}) \hat{m} \gg = |\hat{\beta} \gg | \hat{m} \gg, \quad |(\hat{\beta}) \hat{n} \gg = |\hat{\beta} \gg | \hat{n} \gg, \end{aligned}$$

and the matrix element of the scattering superoperator $\hat{U}(\infty, -\infty)$ is introduced as

$$\begin{aligned} & \ll \hat{n}(\hat{\gamma}) | \hat{U}^{(\pi K)}(\infty, -\infty) | (\hat{\beta}) \hat{m} \gg = \\ & = \frac{1}{2K+1} \sum_Q \ll \hat{n}(\hat{\gamma}) | \hat{U}(\infty, -\infty) | (\hat{\beta}) \hat{m} \gg. \quad (12) \end{aligned}$$

Note as a conclusion, that vectors of the type $|(\hat{\beta}) \hat{m} \gg$ are the special case of the vectors $|\alpha_i j_i (\alpha_f j_f)^+; K_s | \beta_i l_i (\beta_f l_f)^+; K_b | K Q \gg$ which correspond to the momentum coupling scheme

$$\mathbf{j}_i - \mathbf{j}_f = \mathbf{K}_S, \quad \mathbf{l}_i - \mathbf{l}_f = \mathbf{K}_b, \quad \mathbf{K}_S + \mathbf{K}_b = \mathbf{K}, \quad (13)$$

and form a complete set.

3. REPRESENTATION IN THE EXPONENTIAL FORM

There are several possibilities to represent the matrix elements of the scattering matrix in the exponential form. For this purpose we will use the method of a solution of the evolution equation in the matrix form developed in Ref. 12. Since this method allows one to calculate only diagonal matrix elements, the eigenbasis of the matrix of the reduced matrix elements of the superoperator $\hat{U}(\infty, -\infty)$ will be used for calculations. First, transform the formula (11) by expressing the reduced matrix element of the relaxation superoperator $\hat{\Lambda}$ through the diagonal matrix elements of the scattering superoperator. Let $|\hat{\sigma} \gg$ be the eigenvector of the matrix of the reduced matrix elements of the superoperator $\hat{U}(\infty, -\infty)$

$$\hat{U}^{(\pi K)}(\infty, -\infty) | \hat{\sigma}_u \gg = \hat{u} | \hat{\sigma}_u \gg, \quad (14)$$

$$|\hat{\sigma}_u \gg = D | \hat{\kappa}(\hat{\alpha}) \gg. \quad (15)$$

Then instead of Eq. (11) we can write down

$$\begin{aligned} \hat{\Lambda}_{nm}^{(\pi K)} = & -i \eta_b \int dv P(v) \left\{ \delta_{nm}^{\hat{\alpha}} - \sum_{\beta l} \rho_{\beta l}^b \left(\frac{2l+1}{2l+1} \right)^{1/2} \times \right. \\ & \times \sum_{\hat{\kappa}, \hat{\alpha}} \langle\langle \hat{n}(\hat{\gamma}) || D || \hat{\kappa}(\hat{\alpha}) \rangle\rangle \langle\langle \hat{\kappa}(\hat{\alpha}) || D^{-1} || (\hat{\beta}) \hat{m} \rangle\rangle \times \\ & \left. \times \langle\langle \hat{\sigma}_u || \hat{U}^{(\pi K)}(\infty, -\infty) || \hat{\sigma}_u \rangle\rangle \right\}. \end{aligned} \quad (16)$$

Invariance properties of the relaxation superoperator with respect to transformation of the rotation–inversion group have been studied in Ref. 16, where, in particular, it was shown that in the basis appropriate to the momentum coupling scheme given by Eq. (13) its matrix elements are diagonal on K and Q and do not depend on Q . The latter circumstance allows us to calculate the usual matrix element at some fixed value of Q instead of the reduced one. Representing for convenience the interaction Liouvillian $\hat{L}_c(t)$ as a sum of isotropic and anisotropic parts

$$\hat{L}_c(t) = \hat{V}(t) + \hat{R}(t) \quad (17)$$

and applying somewhat changed technique from Ref. 12 for solving the evolution equation, we obtain the reduced matrix element of the scattering superoperator

$$\langle\langle \hat{\sigma}_u || \hat{U}^{(\pi K)}(\infty, -\infty) || \hat{\sigma}_u \rangle\rangle = \exp \left\{ -\langle\langle \hat{\sigma}_u || \hat{A}^{(\pi K)} || \hat{\sigma}_u \rangle\rangle \right\}, \quad (18)$$

where $\hat{A}^{(\pi K)}$ is the matrix of the reduced matrix elements of superoperator \hat{A} which is diagonal in the basis of the vectors $|\alpha_u\rangle$, and \hat{A} is determined by the series

$$\hat{A} = i \int_{-\infty}^{+\infty} \hat{V}(t) dt + \int_{-\infty}^{+\infty} dt_1 \int_{-\infty}^{t_1} dt_2 \hat{L}'(t_1) \hat{L}'(t_2) + \quad (19)$$

+ higher order terms.

Here the prime means that the interaction Liouvillian has no diagonal matrix elements in the basis (15).

Substituting Eq. (18) into Eq. (16) we obtain the required representation for the reduced matrix element of the impact relaxation superoperator

$$\begin{aligned} \hat{\Lambda}_{nm}^{(\pi K)} = & -i \eta_b \int dv P(v) \left\{ \delta_{nm}^{\hat{\alpha}} - \sum_{\beta l} \rho_{\beta l}^b \left(\frac{2l+1}{2l+1} \right)^{1/2} \times \right. \\ & \times \sum_{\hat{\kappa}, \hat{\alpha}} \langle\langle \hat{n}(\hat{\gamma}) || D || \hat{\kappa}(\hat{\alpha}) \rangle\rangle \langle\langle \hat{\kappa}(\hat{\alpha}) || D^{-1} || (\hat{\beta}) \hat{m} \rangle\rangle \times \\ & \left. \times \exp \left\{ -\langle\langle \hat{\sigma}_u || \hat{A}^{(\pi K)} || \hat{\sigma}_u \rangle\rangle \right\} \right\}. \end{aligned} \quad (20)$$

Firstly this result was obtained in Ref. 15. It can be shown that accounting for first two terms in the series (19) the results obtained by iteration method are reproduced exactly after expanding the exponent with an accuracy of the second–order interaction terms. But the formula (20) contains also principal distinctions which will be discussed below.

The formal scheme of calculations of relaxation parameters using Eq. (20) is as follows. The reduced matrix elements of the superoperator \hat{A} are calculated in an initial basis at every step of averaging over classical collision parameters and quantum states of a thermostat particle, thus forming the matrix $\hat{A}^{(\pi K)}$. Then this matrix is reduced to the diagonal form, as a result of which its eigenvalues and the matrix D of eigenvectors became determined. After that the value of the reduced matrix element of the relaxation superoperator is calculated with the use of Eq. (20).

The described scheme apparently is of a high labour capacity, since its every step requires the diagonalization of a matrix which dimension is equal to the product of the line space dimension by the number of states of the buffer molecule involved in the averaging procedure. Therefore, this scheme in its immediate appearance may be applied only in cases when significant simplification is possible, for example, for broadening by particles having no internal structure,¹⁷ or self–broadening, or the ammonia inversion spectrum broadening by foreign gases.^{18–20}

A modification of the scheme which allow the significant enhancement of its potentialities is given in the next part.

Here we note that nondiagonal appearance of the matrix $\hat{A}^{(\pi K)}$ in the initial basis not only leads to emergence of new relaxation parameters, but, as was shown in Refs. 18–20, also affect the spectral line halfwidths, sometimes appreciably.

4. MODIFICATION OF THE FORMULA (20)

If buffer molecule possesses a quantum structure, then, as was mentioned above, the dimension of a matrix to be diagonalized becomes very large. However, in a number of cases interesting for applications it can be reduced down to the dimension of a line space with relatively small losses in accuracy. Let us consider for this purpose the matrix $\hat{A}^{(\pi K)}$. In the second order of a perturbation theory on the interaction Liouvillian it can be represented as a sum of two matrices

$$\begin{aligned} \left\{ \hat{\Lambda}_{nm}^{(\pi K)} \right\} = & \left\{ [i S_1(\hat{n}, \hat{m} | v) + S_2(\hat{n}, \hat{m} | v)_{\text{outer}} + \right. \\ & \left. + \theta_2(\hat{n}, \hat{m} | v)_{\text{middle}}] \delta_{\beta\beta'} \right\} + \left\{ \theta_2(\hat{n}, \hat{m} | v)_{\text{middle}} (1 - \delta_{\beta\beta'}) \right\}, \end{aligned} \quad (21)$$

where the first matrix is diagonal, and the second is nondiagonal in the basis of eigenstates of a buffer molecule.

The explicit expressions for $S_1(\hat{n}, \hat{m} | v)$, $S_2(\hat{n}, \hat{m} | v)_{\text{outer}}$, and $\theta_2(\hat{n}, \hat{m} | v)_{\text{middle}}$ are given in Ref. 2 in a slightly different notations. Considering the second matrix as the perturbation, we represent the matrix of the reduced matrix elements of a scattering superoperator as

$$\begin{aligned} \hat{U}^{(\pi K)}(\infty, -\infty) \approx & \exp \left[-\hat{A}_1^{(\pi K)} \right] \left\{ 1 - \int_0^1 ds \exp \left[\hat{A}_1^{(\pi K)} s \right] \times \right. \\ & \left. \times \hat{A}_2^{(\pi K)} \exp \left[-\hat{A}_1^{(\pi K)} s \right] \right\}. \end{aligned} \quad (22)$$

This formula can be simplified if we assume that the matrices $\hat{A}_1^{(\pi K)}$ and $\hat{A}_2^{(\pi K)}$, the sense of which is evident from Eq. (21), are approximately commutative. In this case

$$\hat{U}^{(\pi K)}(\infty, -\infty) \approx \exp \left[-\hat{A}_1^{(\pi K)} \right] \left(1 - \hat{A}_2^{(\pi K)} \right). \quad (23)$$

Substituting Eq. (23) into Eq. (11), we obtain for an arbitrary reduced matrix element of the impact relaxation operator:

$$\begin{aligned} \hat{\Lambda}_{nm}^{(\pi K)} = & -i \eta_b \int dv P(v) \left\{ \delta_{nm}^{\wedge} - \sum_{\beta l} \rho_{\beta l}^b \sum_{\hat{\kappa}} \sum_{\hat{\gamma}} \left(\frac{2l+1}{2l+1} \right)^{1/2} \times \right. \\ & \times \ll \hat{n}(\hat{\gamma}) || 1 - \hat{A}_2^{(\pi K)} || \hat{\kappa}(\hat{\beta}) \gg \sum_{\hat{r}} \ll \hat{\kappa}(\hat{\beta}) || D || \hat{r}(\hat{\beta}) \gg \times \\ & \left. \times \ll \hat{r}(\hat{\beta}) || D^{-1} || (\hat{\beta}) \hat{m} \gg \exp \{ -\ll \hat{r}(\hat{\beta}) || D \hat{A}_1^{(\pi K)} D || \hat{r}(\hat{\beta}) \gg \} \right\}, \quad (24) \end{aligned}$$

where D is the matrix of eigenvectors of the matrix $\hat{A}_1^{(\pi K)}$ with the dimension equal to that of line space.

The possibility to consider the second matrix in Eq. (21) as a small perturbation follows from the structure of quantities $\theta_2(\hat{n}, \hat{m} | v)_{\text{middle}}$ (see Eq. (36) in Ref. 2). At $\hat{n} = \hat{m}$ these quantities coincide with $S_2(b)_{\text{middle}}$ from Refs. 10 and 11. Their characteristic feature is that, as distinct from $S_2(\hat{n}, \hat{m} | v)_{\text{outer}}$, they do not contain the inner summing over the states of a light-absorbing molecule. Therefore the conditions close to resonance ones are realized rarely, that causes the relatively small values of these quantities.

Let us consider the particular case of isolated spectral lines, when the matrix $\hat{A}^{(\pi K)}$ is obviously diagonal, and, consequently, the matrix D is the unit one. As a result, the formula (24) is simplified significantly

$$\begin{aligned} \hat{\Lambda}_{nm}^{(\pi K)} = & -i \eta_b \int dv P(v) \times \\ & \times \left\{ 1 - \sum_{\beta l} \rho_{\beta l}^b \exp \{ -\ll \hat{m}(\hat{\beta}) || \hat{A}_1^{(\pi K)} || \hat{m}(\hat{\beta}) \gg \} \times \right. \\ & \left. \times \sum_{\hat{\gamma}} \left(\frac{2l+1}{2l+1} \right)^{1/2} [\delta_{\beta \gamma}^{\wedge} - \ll \hat{m}(\hat{\gamma}) || 1 - \hat{A}_2^{(\pi K)} || \hat{m}(\hat{\beta}) \gg] \right\}, \quad (25) \end{aligned}$$

and coincides with the Robert-Bonamy formulas (12) and (13) in Ref. 14 written for spectral line halfwidth and shift, respectively.

5. SELF-BROADENING OF AMMONIA ROTATIONAL SPECTRAL LINES

As an example illustrating the efficiency of the formula (24), we consider self-broadening of rotational lines of ammonia on the basis of the model described in Ref. 21 and shown in Fig. 1. This model is based on the fact that, due to a large value of ammonia rotational constant B , the spectral

exchange between the components of rotational doublets is all that is essential, and consequently, the line space is expanded into a direct sum of two-dimensional subspaces corresponding to the rotational doublets at $K \neq 0$ and to one-dimensional ones corresponding to the singlet lines at $K = 0$. The main details of the calculation are the same as in Ref. 21, and therefore we omit them. The calculations were carried out only for (J, J) doublet lines at $T = 300$ K. The phase effects were not taken into account because of their smallness.²¹

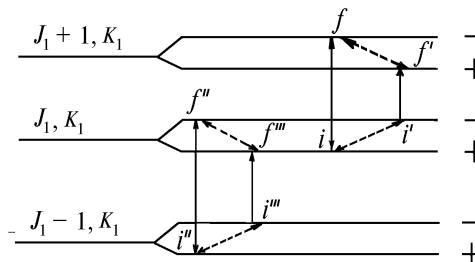


FIG. 1. The model of collisional line interference for the rotational spectrum of ammonia.²¹ The solid arrows correspond to radiative transitions, dashed ones mark collision-induced nonradiative transitions.

The calculational results are presented in Fig. 2, where the experimental data on line halfwidths²² is plotted together with the theoretical data concern both the line halfwidths and the cross-relaxation parameters calculated in Ref. 21 on the basis of Eq. (20) in the assumption that the doublet components are broadened identically which allows the analytical performance of the diagonalization of two-dimensional submatrices of the $\hat{A}^{(\pi K)}(J, K)$ type. Our calculations also did not reveal any noticeable difference in broadening of the doublet components which confirms the arguments given in Ref. 21.

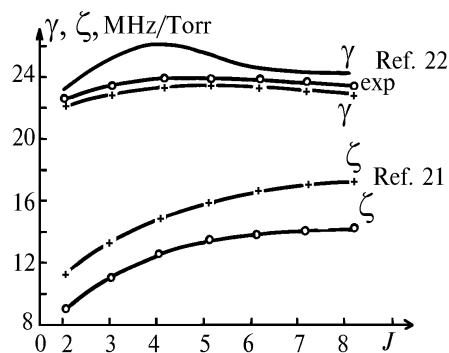


FIG. 2. The line halfwidths and cross-relaxation parameters for rotational (J, J) doublets of the ammonia molecule. The solid line corresponds to the experiment²², $- \circ - \circ -$ corresponds to theoretical data from Ref. 21, and $+ - + -$ corresponds to present calculations.

The formal difference between both the calculations, if neglect the phase effects, thus comes to the presence of the factor $\sum_{\hat{\gamma}} [(2l'+1)(2l+1)]^{1/2} \ll \hat{n}(\hat{\gamma}) || 1 - \hat{A}_2^{(\pi K)} || \hat{\kappa}(\hat{\beta}) \gg$ in Eq. (24) instead of the unit in the correspondent formula in Ref. 21. As a consequence, the values of cross-relaxation parameters obtained in the present calculations increase

approximately up to 20%. As to the line halfwidths, the effect is much weaker and causes their insignificant decrease. These facts are not unexpected and can be seen immediately from analysis of Eq. (24).

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