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The Fokker-Planck Equation of a Laser with Many Modes and Multi-Level Atoms

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Using the Glauber-Sudarshan *P*-representation for the field modes and a quasi-distribution function recently presented for arbitrary quantum systems we derive an exact generalized Fokker-Planck equation for a multi-mode laser containing a set of multi-level atoms with homogeneous and inhomogeneous level broadening. By introduction of suitable collective atomic coordinates this generalized Fokker-Planck equation is reduced to an ordinary one which may serve as a basis for the adequate treatment of laser light statistics.

§ 1. Introduction

After it had been shown theoretically[1] and experimentally[2] that the statistical properties of laser light differ drastically below and above laser threshold a great amount of work has been devoted to a detailed exploration of the statistical properties of laser light[3]. The treatment of photon statistics by Risken[4] and later on by Risken and Vollmer[5], who used a classical Fokker-Planck equation, turned out to be particularly successful and excellent agreement in a region not too far above threshold was found with their predictions[6]. Because the statistics of laser light is caused by quantum mechanical processes the question arose why one is allowed to use the classical Fokker-Planck equation for such a prob-

1 Haken, H.: Z. Physik **181**, 96 (1964).
2 Armstrong, J. A., Smith, A. W.: Phys. Rev. Letters **14**, 68 (1965).
3 For review articles (mainly theoretical) see Haken, H.: Laser theory, vol. XXV/2c of the Encyclopedia of Physics, ed. S. Flügge. Berlin-Heidelberg-New York: Springer 1970. — Risken, H.: Progress in optics, vol. VIII, ed. E. Wolf. Amsterdam: North-Holl. Publ. Comp. 1970. — The articles of Scully, M., Haken, H., Weidlich, W., Louisell, W., Gordon, J. P.: In the Proceedings of the Varenna Summer School "Enrico Fermi", 1967. — Haken, H.: In: Quantum optics, eds. Kay, S. M., Maitland, A. London and New York: Academic Press 1970. — Pike, E. R.: ibd. — The experiments are described in the articles of Arecchi, F. T., Haus, H. A.: Proceedings of the Varenna Summer School "Enrico Fermi", 1967.
4 Risken, H.: Z. Physik **186**, 85 (1965); **191**, 302 (1966).
5 Risken, H., Vollmer, H. D.: Z. Physik **201**, 323 (1967); **204**, 240 (1967).
6 See e.g. Arecchi, F. T., Degiorgio, V., Querzola, K.: Phys. Rev. Letters **20**, 1168 (1967). — Meltzer, D., Mandel, L.: Phys. Rev. Letters **25**, 1151 (1970) and the articles quoted under [3].

lem. Thus there was a challenge to derive Fokker-Planck equations starting from a complete quantum mechanical formulation. The first success was achieved for the damped harmonic oscillator[7]. After it became possible to derive Fokker-Planck-type equations not only for Bose fields but also for 2- and 3-level atoms, Fokker-Planck equations were derived for a laser with a single mode coupled to a system of 2- or 3-level atoms[8]. Finally a generalized Fokker-Planck equation was derived for arbitrary quantum systems, using a formulation particularly well suited for multi-level atoms[9]. All these Fokker-Planck equations apply to quasi-probability distributions.

The purpose of the present paper is to derive a Fokker-Planck equation for a laser with many modes which are coupled to a system of multi-level atoms. Although in a strict sense the generalized Fokker-Planck equation contains arbitrarily high derivatives it is possible to reduce it to an ordinary Fokker-Planck equation containing only the first and second derivative by the introduction of suitable collective excitations of the atoms. The Fokker-Planck equation thus derived may serve as a basis for all calculations of the statistical properties of lasers in multimode action and with atoms having arbitrary numbers of levels.

In subsequent papers by one of us (H.D.V.), the usefulness of the Fokker-Planck equation derived in the present paper will be demonstrated by means of explicit examples.

§ 2. The Density Matrix Equation

We start by writing down the density matrix equation of the whole system. It refers to the quantum states of the individual atoms (first term), the time development of the electromagnetic field (second term) and the coupling of atoms and field due to the interaction Hamiltonian H_{AF}

$$\frac{d\rho}{dt}=\sum_{\mu\varepsilon}\left(\frac{\partial\rho}{\partial t}\right)_{A\mu\varepsilon}+\left(\frac{\partial\rho}{\partial t}\right)_{F}-\frac{i}{\hbar}[H_{AF},\rho]. \tag{2.1}$$

The index μ labels the atoms with respect to their spatial position ξ_μ, the index ε labels them with respect to their different inhomogeneous level broadening. In the following, statistical independence of μ and ε is assumed. For purely homogeneously broadened levels, ε may be dropped.

7 Weidlich, W., Haake, F.: Z. Physik **185**, 30 (1965); **186**, 203 (1965). — Lax, M., Louisell, W. H.: I.E.E.E.J. Quantum Electronics **QE-3**, 47 (1967). — Bonifacio, R., Haake, F.: Z. Physik **200**, 526 (1967).

8 Haken, H., Risken, H., Weidlich, W.: Z. Physik **206**, 355 (1967). — Gordon, J. P.: Phys. Rev. **161**, 367 (1967). — Lax, M., Yuen, H.: Phys. Rev. **173**, 362 (1968).

9 Haken, H.: Z. Physik **219**, 411 (1969).

The first two terms can be calculated by use of a Wangsness-Bloch treatment of atoms and field respectively, coupled to a Markoffian heat reservoir:

a) Atoms. The time development of the density matrix of a n-level atom μ, ε is described by means of the projection operators $P_{ik\mu\varepsilon}$ for the atom μ, ε from level k to level i by the equation

$$\left(\frac{\partial \rho}{\partial t}\right)_{A,\mu,\varepsilon} = \sum_{i,k} \gamma_{ik} P_{ik\mu\varepsilon} \rho P_{ik\mu\varepsilon} + \sum_i (\Gamma_i + i\Omega_{i\varepsilon}) \rho P_{ii\mu\varepsilon} + \sum_i (\Gamma_i - i\Omega_{i\varepsilon}) P_{ii\mu\varepsilon} \rho \tag{2.2}$$

where

$$\Gamma_i = -\tfrac{1}{2} \sum_j \gamma_{ij} .$$

The $P_{ik\mu\varepsilon}$ fulfill the relation

$$P_{ik\mu\varepsilon} P_{i'k'\mu'\varepsilon'} = P_{ik'\mu\varepsilon} \delta_{ki'} \delta_{\mu\mu'} \delta_{\varepsilon\varepsilon'} . \tag{2.3}$$

$\hbar\Omega_{i\varepsilon}$ is the energy of level i. Through its dependence on ε, $\Omega_{i\varepsilon}$ may describe inhomogeneous broadening. The γ_{ik}'s describe for $i \neq k$ the transition rates from level i to level k, for $i=k$ the level width caused by phase destroying processes occurring in level i.

The explicit form of Eq. (2.2) is taken from Schmid and Risken[10] where the transformation from the projection operator description to the density matrix formulation had been made.

b) Field. The time development of the electromagnetic field within the cavity of volume V, written in terms of the field creation and annihilation operators b_λ^+, b_λ for each mode λ, is given by[11]

$$\left(\frac{\partial \rho}{\partial t}\right)_F = \sum_\lambda \{ -i\nu_\lambda [b_\lambda^+ b_\lambda, \rho] - \kappa_\lambda \{ [b_\lambda^+, b_\lambda \rho] + [\rho b_\lambda^+, b_\lambda] \} + 2\kappa_\lambda n_{\lambda\,\mathrm{th}} [[b_\lambda, \rho], b_\lambda^+] \} \tag{2.4}$$

where $[A, B] = AB - BA$.

The commutation relations for b_λ^+, b_λ are

$$[b_\lambda, b_{\lambda'}^+] = \delta_{\lambda,\lambda'}, \quad [b_\lambda, b_{\lambda'}] = [b_\lambda^+, b_{\lambda'}^+] = 0 . \tag{2.5}$$

$n_{\lambda\,\mathrm{th}} = (\exp(\hbar\nu_\lambda / kT) - 1)^{-1}$ is the mean number of thermal quanta of the mode λ in the cavity of temperature T, ν_λ is the eigenfrequency of mode λ and κ_λ the corresponding damping constant.

The third term describes the (coherent) *interaction.*

The linear interaction of field and atoms can be derived by use of the Hamiltonian

$$H_{AF} = \hbar \sum_{i,k,\lambda',\mu,\varepsilon} g_{ik} h_{\mu\lambda'} P_{ik\mu\varepsilon} (b_{\lambda'}^+ + b_{\lambda'}) . \tag{2.6}$$

10 Schmid, C., Risken, H.: Z. Physik **189**, 365 (1966).
11 Weidlich, W., Haake, F.: See Ref. [7].

The coupling constant $g_{ik}h_{\mu\lambda'}$ for the transition between the levels i and k of atom μ, ε and the electric field of mode λ' is given in the dipole approximation by (standing electromagnetic waves[12])

$$g_{ij}h_{\mu\lambda'} = g_{ij}h_{\lambda'}\sin k_{\lambda'}\xi_\mu = -\frac{2e}{m}\sqrt{\frac{\pi}{\hbar\omega_{\lambda'}V}}\sin(k_{\lambda'}\xi_\mu)\cdot\int\varphi_i^*(\underset{\sim}{e}_{\lambda'}\cdot\underset{\sim}{p})\varphi_j\,d\tau \quad (2.7)$$

and can therefore be split up into a field and an atomic part. It should be noted that in general $g_{ii}=0$ because of the definite parity of the atomic wave functions.

Using the explicit expressions (2.2), (2.4), (2.6), the density matrix equation reads[13]

$$\begin{aligned}\frac{\partial\rho}{\partial t} = \sum_\lambda \{ & -i\nu_\lambda(b_\lambda^+ b_\lambda\rho - \rho b_\lambda^+ b_\lambda) \\ & -\kappa_\lambda(b_\lambda^+ b_\lambda\rho + \rho b_\lambda^+ b_\lambda - 2b_\lambda\rho b_\lambda^+) \\ & +2\kappa_\lambda n_{\lambda\,\mathrm{th}}(b_\lambda^+\rho b_\lambda + b_\lambda\rho b_\lambda^+ - b_\lambda^+ b_\lambda\rho - \rho b_\lambda b_\lambda^+)\} \\ & + \sum_{i,k,\mu,\varepsilon}\gamma_{ik}P_{ki\mu\varepsilon}\rho P_{ik\mu\varepsilon} \\ & + \sum_{i,k,\lambda,\mu,\varepsilon}[(\Gamma_i - i\Omega_{i\varepsilon})\delta_{ik}\delta_{\lambda,0} - i g_{ik}h_{\mu\lambda}(b_\lambda^+ + b_\lambda)]P_{ik\mu\varepsilon}\rho \\ & + \sum_{i,k,\lambda,\mu,\varepsilon}\rho P_{ik\mu\varepsilon}[(\Gamma_i + i\Omega_{i\varepsilon})\delta_{ik}\delta_{\lambda,0} + i g_{ik}h_{\mu\lambda}(b_\lambda^+ + b_\lambda)]. \end{aligned} \quad (2.8)$$

§ 3. Definition of the Distribution Function, Calculation of Expectation Values

The distribution function f of the variables u, u^* and v, corresponding to the operators b^+, b and P of field and atoms, is defined as usual by use of the Fourier transform f of the characteristic function F.

$$\begin{aligned} f(\{u\},\{v\},t) = \mathfrak{N}\int\cdots\int & e^{-\sum_{i,k,\mu,\varepsilon} x_{ik\mu\varepsilon}v_{ik\mu\varepsilon} - \sum_\lambda (y_\lambda u_\lambda + y_\lambda^* u_\lambda^*)} \\ & \cdot F(\{x\},\{y\},t)\,d\{x\}\,d\{y\}. \end{aligned} \quad (3.1)$$

$\mathfrak{N}$ means the normalization constant, y_λ, y_λ^* and $x_{ik\mu\varepsilon}$ are the classical variables related to operators b_λ, b_λ^+ and $P_{ik\mu\varepsilon}$, respectively*. The variables y_λ, y_λ^*, $x_{ik\mu\varepsilon}$ are imaginary, so that (3.1) is indeed a Fourier

* Note that * does not mean here the complex conjugate but denotes another set of variables, related to the others by

$$u^* = \bar{u}, \quad u = \bar{u}^*, \quad \bar{y} = -y^*, \quad \bar{y}^* = -y.$$

The bar indicates the complex conjugate.

12 See e.g. Haken, H.: Z. Physik **190**, 327 (1966).

13 See e.g. the first article quoted under Ref. [3].

transform. The characteristic function F is derived from the density matrix $\rho(t)$ by taking the trace over the whole system.

$$F=\mathrm{Tr}(O\rho) \tag{3.2}$$

after multiplying it with an operator O that can be chosen suitably. In order to arrive at a definition of the distribution function f, which also holds classically, (3.2) must be a Fourier transform from ρ to F because ρ is known to be the distribution function. The classical function O must therefore be a product of exponentials. The quantum operator O is similarly composed of exponential operators whose sequence can be suitably chosen.

In a previous paper[14] we have introduced such an operator O and given rules how operators applied to O act on the characteristic function F. While the former treatment is most suitable for a single n-level atom, it is advisable to define O for a system of atoms in the following generalized form.

$$\begin{aligned} O(\{x\},\{y\}) &= \prod_{\mu,\varepsilon} O_{\mu\varepsilon} \cdot \prod_{\lambda} O_{\lambda} \\ &= \prod_{\mu,\varepsilon} \Big\{ \prod_{i<k} e^{x_{ik\mu\varepsilon} P_{ik\mu\varepsilon}} \prod_{i} e^{x_{ii\mu\varepsilon} P_{ii\mu\varepsilon}} \prod_{i>k} e^{x_{ik\mu\varepsilon} P_{ik\mu\varepsilon}} \Big\} \\ &\quad \cdot \prod_{\lambda} \{ e^{y_\lambda^* b_\lambda^+} \cdot e^{y_\lambda b_\lambda} \}. \end{aligned} \tag{3.3}$$

The operators b occur in the same way as in Glaubers P-representation[15].

In our further calculation it is rather unimportant whether differential equations for F or f are derived. Due to the Fourier transform (3.1) the characteristic and the distribution function are connected by the relation

$$x^q \left(\frac{d}{dx}\right)^p F(x) \leftrightarrow \left(-\frac{\partial}{\partial v}\right)^q v^p f(v). \tag{3.4}$$

In order to be able to transform the operator equation (2.8) into a partial differential equation with the variables v, u or x, y for the function f or F, we must know how operators acting on O can be replaced by differential operators. These relations can be derived by use of the commutators. For the field these relations read[16]

$$\begin{aligned} b_\lambda^+ O_\lambda &= \frac{\partial O_\lambda}{\partial y_\lambda^*}; & b_\lambda O_\lambda &= \left(\frac{\partial}{\partial y_\lambda} + y_\lambda^*\right) O_\lambda \\ O_\lambda b_\lambda^+ &= \left(\frac{\partial}{\partial y_\lambda^*} + y_\lambda\right) O_\lambda; & O_\lambda b_\lambda &= \frac{\partial}{\partial y_\lambda} O_\lambda. \end{aligned} \tag{3.5}$$

14 See Ref. [9].

15 Compare the paper of Lax, M., Louisell, W. H., Ref. [7].

16 See Ref. [7, 8].

For the projection operators of atoms the corresponding relations have been determined previously and read[17]

$$\begin{aligned} O_{\mu\varepsilon} P_{ik\mu\varepsilon} &= \sum_{l,m} N^{(1)}_{iklm}(\{x\}_{\mu\varepsilon}) \frac{\partial}{\partial x_{lm\mu\varepsilon}} O_{\mu\varepsilon} \\ P_{ik\mu\varepsilon} O_{\mu\varepsilon} &= \sum_{l,m} N^{(2)}_{iklm}(\{x\}_{\mu\varepsilon}) \frac{\partial}{\partial x_{lm\mu\varepsilon}} O_{\mu\varepsilon} \\ P_{ik\mu\varepsilon} O_{\mu\varepsilon} P_{ki} &= \sum_{l,m} M_{ikki,lm}(\{x\}_{\mu\varepsilon}) \frac{\partial}{\partial x_{lm\mu\varepsilon}} O_{\mu\varepsilon}. \end{aligned} \tag{3.6}$$

The functions M and N are given explicitly in the Appendix. The argument $\{x\}_{\mu\varepsilon}$ stands for all $x_{l'm'\mu\varepsilon}$ with fixed μ and ε, but variable l' and m' (compare the Appendix).

In order to derive a differential equation for the distribution function we multiply the density matrix equation on both sides from the left with O and take the trace over the whole system. The further steps have been described previously[18] and lead to the equation

$$\begin{aligned} \frac{\partial F}{\partial t} = \sum_{\lambda} \Big\{ & i\nu_\lambda \left(y^*_\lambda \frac{\partial}{\partial y^*_\lambda} - y_\lambda \frac{\partial}{\partial y_\lambda} \right) \\ & - \kappa_\lambda \left(y^*_\lambda \frac{\partial}{\partial y^*_\lambda} + y_\lambda \frac{\partial}{\partial y_\lambda} \right) + 2\kappa_\lambda n_{\lambda\,\mathrm{th}} y^*_\lambda y_\lambda \Big\} F \\ & + \sum_{i,k,l,m,\mu,\varepsilon} \gamma_{ik} M_{ikki,lm}(\{x\}_{\mu\varepsilon}) \frac{\partial}{\partial x_{lm\mu\varepsilon}} F \\ & + \sum_{i,l,m,\mu,\varepsilon} i\Omega_{i\varepsilon} (N^{(2)}_{iilm}(\{x\}_{\mu\varepsilon}) - N^{(1)}_{iilm}(\{x\}_{\mu\varepsilon})) \frac{\partial}{\partial x_{lm\mu\varepsilon}} F \\ & + \sum_{i,k,l,m,\mu,\varepsilon} (-\tfrac{1}{2}\gamma_{ik}) (N^{(2)}_{iilm}(\{x\}_{\mu\varepsilon}) + N^{(1)}_{iilm}(\{x\}_{\mu\varepsilon})) \frac{\partial}{\partial x_{lm\mu\varepsilon}} F \\ & + \sum_{i,k,l,m,\lambda,\mu,\varepsilon} i g_{ik} h_{\mu\lambda} (N^{(2)}_{iklm}(\{x\}_{\mu\varepsilon}) - N^{(1)}_{iklm}(\{x\}_{\mu\varepsilon})) \\ & \qquad \cdot \left(\frac{\partial}{\partial y^*_\lambda} + \frac{\partial}{\partial y_\lambda} \right) \frac{\partial}{\partial x_{lm\mu\varepsilon}} F \\ & + \sum_{i,k,l,m,\lambda,\mu,\varepsilon} i g_{ik} h_{\mu\lambda} (N^{(2)}_{iklm}(\{x\}_{\mu\varepsilon}) y^*_\lambda - N^{(1)}_{iklm}(\{x\}_{\mu\varepsilon}) y_\lambda) \frac{\partial}{\partial x_{lm\mu\varepsilon}} F. \end{aligned} \tag{3.7}$$

The first sum describes the lightfield, the second, third and fourth the atomic motion. The fifth and sixth sum refer to the coupling between field and atoms. This differential equation for the characteristic function is of very high order, namely of the order $N \cdot n^2 + 2M$ where N is the

17 See Ref. [9].
18 See Ref. [8].

number of atoms, n that of atomic levels and M the number of field modes under consideration. It is known from other work that the laser dynamics can be very often described by much less variables, if a suitable set of macroscopic variables is used. For instance in a two level system with only one mode, those adequate collective variables are the total atomic inversion and the total dipole moment[19]. This suggests to try to introduce similar collective variables for our multimode and multilevel system. On account of the boundary condition one is led to introduce 2 types of collective operators in the definition of the distribution function: Those which are connected with the off-diagonal atomic matrix elements, which may be decomposed using a sine wave and those connected with the diagonal elements corresponding to atomic occupation numbers which are decomposed into cosine waves. In order to treat those two cases simultaneously we introduce the following decomposition and its inverse

$$p_{ik\lambda\varepsilon}=\sum_{|\mu|} C_{ik\lambda\mu} P_{ik\mu\varepsilon}; \qquad P_{ik\mu\varepsilon}=\frac{1}{N}\sum_{|\lambda|} C^*_{ik\lambda\mu}\, p_{ik\lambda\varepsilon} \tag{3.8}$$

where the coefficients C are defined by

$$C_{ik\lambda\mu}=\delta_{ik}\cos k_\lambda \xi_\mu - i(1-\delta_{ik})\sin k_\lambda \xi_\mu . \tag{3.9}$$

In the definition of F and f both operators and classical variables occur. It is advisable to introduce collective variables for both sets of quantities. We first introduce collective *operators*. Due to the symmetry relation

$$P_{ik\mu}=(2\delta_{ik}-1)P_{ik-\mu}; \qquad p_{ik\lambda}=(2\delta_{ik}-1)\,p_{ik-\lambda} \tag{3.10}$$

the sums in (3.8) have to run over all $|\mu|$ and $|\lambda|$ respectively, whereas the sign can be chosen arbitrarily for each λ, μ. (The index ε has been dropped in (3.10).)

The corresponding collective coordinates (the classical variables) are defined in such a way that sums in the exponents of the function O, (3.3), remain invariant under the transformation

$$\sum_{|\lambda|} p_{ik\lambda\varepsilon} X_{ik\lambda\varepsilon}=\sum_{|\mu|} P_{ik\mu\varepsilon}\, x_{ik\mu\varepsilon}. \tag{3.11}$$

This leads after a short transformation to the following definition and its inverse

$$\begin{aligned} x_{ik\mu\varepsilon}&=\sum_{|\lambda|} C_{ik\lambda\mu} X_{ik\lambda\varepsilon} \\ X_{ik\lambda\varepsilon}&=\frac{1}{N}\sum_{|\mu|} C^*_{ik\lambda\mu}\, x_{ik\mu\varepsilon}. \end{aligned} \tag{3.12}$$

19 See Ref. [1].

The transformation of the derivatives is given by

$$\frac{\partial}{\partial x_{ik\mu\varepsilon}}=\frac{1}{N}\sum_{|\lambda|}C^*_{ik\lambda\mu}\frac{\partial}{\partial X_{ik\lambda\varepsilon}}. \tag{3.13}$$

For the Fourier transformed variables we find in complete analogy to (3.8) the relations

$$V_{ik\lambda\varepsilon}=\sum_{|\mu|}C_{ik\lambda\mu}v_{ik\mu\varepsilon};\qquad v_{ik\mu\varepsilon}=\frac{1}{N}\sum_{|\lambda|}C^*_{ik\lambda\mu}V_{ik\lambda\varepsilon}. \tag{3.14}$$

A simple inspection of the functional determinant reveals that it is a constant for this transformation so that we can immediately proceed to the definition of the distribution function by means of the collective variables:

$$\begin{aligned}f(\{V\},\{u\})=\mathfrak{N}'\int\cdots\int e^{-\sum_{i,k,\lambda,\varepsilon}X_{ik\lambda\varepsilon}V_{ik\lambda\varepsilon}-\sum_{\lambda}(y_\lambda u_\lambda+y^*_\lambda u^*_\lambda)}\\ \cdot F(\{X\},\{y\})\,d\{X\}\,d\{y\}.\end{aligned} \tag{3.15}$$

§ 4. Differential Equation of the Characteristic Function in Collective Coordinates

The main task is now to transform the Eq. (3.7), which had referred to individual coordinates, to an equation referring to the collective coordinates introduced above. We skip the elementary transformation and write down immediately the differential equation

$$\begin{aligned}\frac{\partial F}{\partial t}=&\sum_{|\lambda|}\left\{iv_\lambda\left(y^*_\lambda\frac{\partial}{\partial y^*_\lambda}-y_\lambda\frac{\partial}{\partial y_\lambda}\right)\right.\\ &\left.-\kappa_\lambda\left(y^*\frac{\partial}{\partial y^*_\lambda}+y_\lambda\frac{\partial}{\partial y_\lambda}\right)+2\kappa_\lambda n_{\lambda\,\mathrm{th}}y^*_\lambda y_\lambda\right\}F\\ &+\sum_{l,m,|\lambda|,\varepsilon}K_{lm\lambda\varepsilon}(\{X\}_\varepsilon)\frac{\partial}{\partial X_{lm\lambda\varepsilon}}F\\ &+\sum_{\substack{l,m\\|\lambda|,|\lambda'|,\varepsilon}}\left(J^{(2)}_{lm\lambda\lambda'}(\{X\}_\varepsilon)-J^{(1)}_{lm\lambda\lambda'}(\{X\}_\varepsilon)\right)\left(\frac{\partial}{\partial y_{\lambda'}}+\frac{\partial}{\partial y^*_{\lambda'}}\right)\frac{\partial}{\partial X_{lm\lambda\varepsilon}}\cdot F\\ &+\sum_{\substack{l,m\\|\lambda|,|\lambda'|,\varepsilon}}\left(J^{(2)}_{lm\lambda\lambda'}(\{X\}_\varepsilon)\,y^*_{\lambda'}-J^{(1)}_{lm\lambda\lambda'}(\{X\}_\varepsilon)\,y_{\lambda'}\right)\frac{\partial}{\partial X_{lm\lambda\varepsilon}}\cdot F\end{aligned} \tag{4.1}$$

where the following abbreviations were used

$$J^{(2)}_{lm\lambda\lambda'}(\{X\}_\varepsilon)=\sum_{i,k}\frac{1}{N}\cdot g_{ik}h_{\lambda'}\,i\sum_{|\mu|}C^*_{lm\lambda\mu}N^{(2)}_{(1)ikl\,m}(\{x\}_{\mu\varepsilon})\cdot\sin(k_{\lambda'}\xi_\mu) \tag{4.2}$$

and

$$K_{lm\lambda\varepsilon}(\{X\}_\varepsilon)=\sum_{i,k}\gamma_{ik}\cdot\frac{1}{N}\sum_{|\mu|}C^*_{lm\lambda\mu}\{M_{ikki,lm}(\{x\}_{\mu\varepsilon})$$
$$-\tfrac{1}{2}(N^{(2)}_{iilm}(\{x\}_{\mu\varepsilon})+N^{(1)}_{iilm}(\{x\}_{\mu\varepsilon}))\} \tag{4.3}$$
$$+\sum_{i,k}\delta_{ik}\cdot\frac{1}{N}\sum_{|\mu|}i\,\Omega_{i\varepsilon}\cdot(N^{(2)}_{iilm}(\{x\}_{\mu\varepsilon})-N^{(1)}_{iilm}(\{x\}_{\mu\varepsilon}))\,C^*_{lm\lambda\mu}.$$

$\{X\}_\varepsilon$ stands for the set of variables $X_{l'm'\lambda\varepsilon}$ for all possible l', m', λ, but fixed ε.

The functions N_{iklm} and $M_{ikki,lm}$ on the right hand side are to be taken as the functions defined in the Appendix. In these functions the individual atomic variables are to be expressed by the collective ones according to the formulas (3.12). The equation for F is the most general result of our present analysis. If we assume that the original density matrix equation is exact this new equation describes exactly the time dependent and stationary behaviour of a laser with many modes and a complete system of atomic variables. Of course, for practical purposes this equation is still far too general, but we want to show that there are several effective ways to cut down the number of variables. Usually the number of atoms in a laser is very large. Because the collective variables $V_{ik\lambda\varepsilon}$ increase with the number of atoms we may anticipate that the corresponding relative fluctuations will decrease. On account of the transformation (3.4) we expect the variables $X_{ik\lambda\varepsilon}$ and y_λ to become smaller with increasing atomic number. Thus we may try to expand the coefficients $J_{lm\lambda\lambda'}$ and $K_{lm\lambda\varepsilon}$ into a power series of the number of atoms, N. Because we are dealing with a differential equation, the order of magnitude of terms containing derivatives can be determined only if the distribution function is known or is known afterwards so that we can make our assumptions selfconsistent. Indeed, all examples calculated so far, show that f is a smooth function, in which the field distribution depends on the parameter N in a characteristic manner used below. Therefore we are allowed to compare the coefficients and the derivatives in an adequate way.

In order to do the investigation with respect to the variables $X_{ik\lambda\varepsilon}$ and y_λ we remind the reader of the connection (3.4) between the variables and their Fourier transform $V_{ik\lambda}$ and u_λ. For the Fourier transformation a factor is to be replaced by a derivative and vice versa according to

$$X_{ik\lambda\varepsilon}\to-\frac{\partial}{\partial V_{ik\lambda\varepsilon}},\qquad \frac{\partial}{\partial X_{ik\lambda'\varepsilon}}\to V_{ik\lambda\varepsilon},$$
$$y_\lambda\to-\frac{\partial}{\partial u_\lambda},\qquad \frac{\partial}{\partial y_\lambda}\to u_\lambda. \tag{4.4}$$

Table. *Dependence of the terms in Eq.* (4.1) *on the number of atoms within the resonator when the coefficients* $N^{(1)}$, $N^{(2)}$, M *are expanded into a power series of the coordinates*

Power of the dependence on N of the following terms		Region relative to threshold		
		above	at	below
$X_{l i \lambda \varepsilon}$		-1	-1	-1
g_{ik}		$-1/2$	$-1/2$	$-1/2$
$X_{i j \lambda \varepsilon}$ $(i \neq j)$		-1	$-3/4$	$-1/2$
y_λ		$-1/2$	$-1/4$	0
Field part of (4.1)		0, -1	0, $-1/2$	0
atomic part, diagonal	0	1	1	1
$K_{l l \lambda}(\partial/\partial X_{l l \lambda \varepsilon})$	1	0	0	0
power of X in K	2	-1	$-1/2, -1$	$0, -1$
	3	-2	$-3/2, -2$	$-1, -2$
atomic part, off-diagonal	1	0	0	0
$K_{l m \lambda}(X)$ $(\partial/\partial X_{l m \lambda \varepsilon})$	2	-1	-1	-1
power of X in K	3	-2	$-3/2, -2$	$-1, -2$
Interaction, diagonal part				
$J_{l l \lambda \lambda'}(X)\left(\frac{\partial}{\partial y_{\lambda'}} + \frac{\partial}{\partial y^*_{\lambda'}}\right)\frac{\partial}{\partial X_{l l \lambda \varepsilon}}$	0	—	—	—
	1	0	0	0
power of X in J	2	-1	-1	-1
interaction, off-diagonal part $(l \neq m)$				
$J_{l m \lambda \lambda'}(X)\left(\frac{\partial}{\partial y_{\lambda'}} + \frac{\partial}{\partial y^*_{\lambda'}}\right)\frac{\partial}{\partial X_{l m \lambda \varepsilon}}$	0	1	1/2	0
	1	0	$-1/2$	-1
power of X in J	2	-1	$-1, -3/2$	$-1, -2$
interaction, diagonal part				
$J_{l l \lambda \lambda'} y^*_{\lambda'} \frac{\partial}{\partial X_{l l \lambda \varepsilon}}$	0	—	—	—
	1	-1	$-1/2$	0
power of X in J	2	-2	$-3/2$	-1
interaction, off-diagonal part				
$J_{l m \lambda \lambda'} y^*_{\lambda'} \frac{\partial}{\partial X_{l m \lambda \varepsilon}}$ $(l \neq m)$	0	0	0	0
	1	-1	-1	-1
power of X in J	2	-2	$-3/2, -2$	$-1, -2$

Consequently the dependence of the variables $X_{ik\lambda\varepsilon}$ and $V_{ik\lambda}$ on N is reciprocal. In the Table the powers of N of the corresponding contributions are given for 3 regions, above, at and below threshold. In the Table we assume that the field amplitude below threshold is independent of the number of atoms but above threshold depends linearly on the number of atoms. This assumption is substantiated by the detailed treatment of a single mode laser and is selfconsistent in this treatment.

The results of the Table can be interpreted as follows: At and below threshold it is sufficient to retain terms up to second order for the atomic part and up to first order in the interaction term. Thus one obtains an equation whose Fourier transform contains derivations only up to second order so that the usual Fokker-Planck equation results. Far above threshold the interaction term must be treated in the same way as the atomic part up to second order. In this way we obtain a range of validity which goes beyond that of the Fokker-Planck equations derived so far. The criteria of how far we have to go in our expansion is the following. We retain the field term as the exact one and expand the other terms in powers of the atomic number at least to the same order. Using the results of the Table we expand the coefficients $N^{(1)}_{iklm}$, $N^{(2)}_{iklm}$, $M_{ikki,lm}$ as given in the Appendix and evaluate the sums over the atomic indices as far as needed.

This leads us to the expression

$$
\begin{aligned}
K_{lm\lambda\varepsilon}(\{X\}_\varepsilon) = {} & \delta_{lm}\sum_{i,k}(\gamma_{ki}\delta_{lk}-\gamma_{ik}\delta_{il})X_{ii\lambda\varepsilon} \\
& +\left[i(\Omega_{l\varepsilon}-\Omega_{m\varepsilon})-\tfrac{1}{2}(1-\delta_{lm})\sum_k(\gamma_{lk}+\gamma_{mk})\right]X_{lm\lambda\varepsilon} \\
& +\tfrac{1}{2}\delta_{lm}\sum_{\substack{i,k,p\\ |\lambda'|,|\lambda''|}}\left[\gamma_{lp}\delta_{ik}(\delta_{ip}+\delta_{il})-2\gamma_{ik}\delta_{li}\delta_{ip}\right] \\
& \qquad \cdot X_{ii\lambda'\varepsilon}X_{kk\lambda''\varepsilon}\delta_{\lambda+\lambda'+\lambda'',0} \\
& +\sum_{k,|\lambda'|,|\lambda''|}\left[\delta_{m<l}(\gamma_{lk}-\delta_{kl}\sum_i\gamma_{li})+\delta_{l<m}(\gamma_{mk}-\delta_{mk}\sum_i\gamma_{mi})\right] \\
& \qquad \cdot X_{kk\lambda'\varepsilon}X_{lm\lambda''\varepsilon}\delta_{\lambda+\lambda'+\lambda'',0} \qquad (4.5) \\
& +\delta_{lm}\sum_{\substack{i,k\\ |\lambda'|,|\lambda''|}}\left(\gamma_{lk}\delta_{k<i}+\sum_p\gamma_{kp}\delta_{l<k}\delta_{il}\right) \\
& \qquad \cdot X_{ik\lambda'\varepsilon}X_{ki\lambda''\varepsilon}\delta_{\lambda+\lambda'+\lambda'',0}\,.
\end{aligned}
$$

In a similar way the somewhat lengthy but elementary calculation yields for $J^{(1)}$ and $J^{(2)}$ which occur in (4.1):

$$
\begin{aligned}
J^{(1)}_{lm\lambda\lambda'}(\{X\}_\varepsilon) = {} & -h_{\lambda'}g_{lm}\delta_{\lambda,\lambda'} \\
& +h_{\lambda'}g_{lm}\delta_{l<m}\sum_{i,|\lambda''|}(\delta_{il}-\delta_{im})X_{ii\lambda''\varepsilon}\delta_{\lambda+\lambda'+\lambda'',0} \\
& +h_{\lambda'}\delta_{lm}\sum_{i,k,|\lambda''|}g_{ik}\delta_{i<k}(\delta_{mi}-\delta_{kl})X_{ki\lambda''\varepsilon}\delta_{\lambda+\lambda'+\lambda'',0} \\
& +\tfrac{1}{2}h_{\lambda'}g_{lm}\delta_{l<m}\sum_{i,k,|\lambda''|,|\lambda'''|}\left[\delta_{ik}(\delta_{il}+\delta_{im})\right. \\
& \qquad \left.-(\delta_{il}\delta_{km}+\delta_{kl}\delta_{im})\right] \\
& \qquad \cdot X_{ii\lambda''\varepsilon}X_{kk\lambda'''\varepsilon}\delta_{\lambda+\lambda'+\lambda''+\lambda''',0} \qquad (4.6)
\end{aligned}
$$

$$+\sum_{\substack{i,k,p,q\\|\lambda''|,|\lambda'''|}} h_{\lambda'}\big[-g_{ik}\big(\delta_{i<l}\delta_{l<k}\delta_{l<m}(1-\delta_{km})$$
$$+(\delta_{im}+\delta_{i<m})\,\delta_{m<k}\delta_{m<l}\big)\delta_{lp}\delta_{mq}$$
$$+g_{lk}\delta_{pq}\delta_{im}\delta_{l<p}\delta_{p<k}\delta_{p<m}$$
$$+g_{im}(1-\delta_{lm})\,\delta_{i<m}(\delta_{pm}+\delta_{p<m})$$
$$\cdot\,\delta_{pq}\delta_{lk}\delta_{i<p}\delta_{p<l}\big]$$
$$\cdot\,X_{pi\lambda''\varepsilon}X_{kq\lambda'''\varepsilon}\delta_{\lambda+\lambda'+\lambda''+\lambda''',0}\,.$$

$J^{(2)}_{lm\lambda\lambda'}$ may be obtained from $J^{(1)}_{lm\lambda\lambda'}$ by the following relations

$$\begin{array}{ccccc} J^{(1)}_{lm\lambda\lambda'} & l & m & g_{ik} \\ | & | & | & | \\ J^{(2)}_{lm\lambda\lambda'} & m & l & g_{ki}\,. \end{array} \tag{4.7}$$

After insertion of $K_{lm\lambda\varepsilon}$, $J^{(1)}_{lm\lambda\lambda'}$ and the corresponding relation for $J^{(2)}_{lm\lambda\lambda'}$ we obtain a differential equation for the characteristic function. Our final goal is to find the ordinary Fokker-Planck equation for the distribution function itself which may be immediately obtained from that for the characteristic function by taking the Fourier transform. Our final result reads

$$\frac{\partial f}{\partial t}=\sum_{|\lambda|}\frac{\partial}{\partial u^*_\lambda}\{(-iv_\lambda+\kappa_\lambda)u^*_\lambda+h_\lambda\sum_{l,m,\varepsilon}g_{lm}V_{lm\lambda\varepsilon}\}f$$
$$+\sum_{|\lambda|}\frac{\partial}{\partial u_\lambda}\{(iv_\lambda+\kappa_\lambda)u_\lambda-h_\lambda\sum_{l,m,\varepsilon}g_{lm}V_{lm\lambda\varepsilon}\}f$$
$$+\sum_{i,|\lambda|,\varepsilon}\frac{\partial}{\partial V_{ii\lambda\varepsilon}}\{\sum_k(\gamma_{ik}V_{ii\lambda\varepsilon}-\gamma_{ki}V_{kk\lambda\varepsilon})+\sum_{k,|\lambda'|\,|\lambda''|}h_{\lambda'}(u_{\lambda'}+u^*_{\lambda'})$$
$$\cdot\,(g_{ik}V_{ik\lambda''\varepsilon}-g_{ki}V_{ki\lambda''\varepsilon})\delta_{\lambda+\lambda'+\lambda'',0}\}f$$
$$+\sum_{i,k,|\lambda|,\varepsilon}\frac{\partial}{\partial V_{ik\lambda\varepsilon}}\{[i(\Omega_{k\varepsilon}-\Omega_{i\varepsilon})+\tfrac{1}{2}(1-\delta_{ik})\sum_l(\gamma_{il}+\gamma_{kl})]V_{ik\lambda\varepsilon}$$
$$+\sum_{|\lambda'|\,|\lambda''|}h_{\lambda'}g_{ki}(u_{\lambda'}+u^*_{\lambda'})(V_{kk\lambda''\varepsilon}-V_{ii\lambda''\varepsilon})\delta_{\lambda+\lambda'+\lambda'',0}\}f$$
$$+\sum_{|\lambda|\varepsilon}\frac{\partial^2}{\partial u_\lambda\partial u^*_\lambda}\{2k_\lambda n_{\lambda\mathrm{th}}\}f$$
$$+\sum_{\substack{i,k,l\\|\lambda|,|\lambda'|,|\lambda''|\varepsilon}}\frac{\partial^2}{\partial V_{ii\lambda\varepsilon}\partial V_{kk\lambda'\varepsilon}}\{[\tfrac{1}{2}\delta_{ik}(\gamma_{li}V_{ll\lambda''\varepsilon}+\gamma_{il}V_{ii\lambda''\varepsilon})$$
$$-\gamma_{ik}\delta_{li}V_{ii\lambda''\varepsilon}]\delta_{\lambda+\lambda'+\lambda'',0}$$
$$-\tfrac{1}{2}h_{\lambda''}(u_{\lambda''}+u^*_{\lambda''})(\delta_{ik}-\delta_{kl})(\delta_{i<l}-\delta_{l<i})$$
$$\cdot\sum_{|\lambda'''|}(g_{il}V_{il\lambda'''\varepsilon}-g_{li}V_{li\lambda'''\varepsilon})\delta_{\lambda+\lambda'+\lambda''+\lambda''',0}\}f$$

$$
\begin{aligned}
&+\sum_{\substack{i,l,m\\ |\lambda|,|\lambda'|,|\lambda''|\varepsilon}} \frac{\partial^2}{\partial V_{ii\lambda\varepsilon}\partial V_{lm\lambda'\varepsilon}}\{[(\delta_{m<l}\gamma_{li}+\delta_{l<m}\gamma_{mi})\\
&\quad -\sum_k \gamma_{ik}(\delta_{m<l}\delta_{il}+\delta_{l<m}\delta_{im})]\,V_{lm\lambda''\varepsilon}\,\delta_{\lambda+\lambda'+\lambda'',0}\}f\\
&+\sum_{\substack{i,k,l,m,p\\ |\lambda|,|\lambda'|,|\lambda''|\varepsilon}} \frac{\partial^2}{\partial V_{li\lambda\varepsilon}\partial V_{km\lambda'\varepsilon}}\{\delta_{ik}\delta_{lm}(\delta_{l<i}\gamma_{pl}V_{pp\lambda''\varepsilon} \qquad (4.8)\\
&\quad +\delta_{i<l}\gamma_{lp}V_{ii\lambda''\varepsilon})\,\delta_{\lambda+\lambda'+\lambda'',0}+h_{\lambda''}(u_{\lambda''}+u^*_{\lambda''})\\
&\quad \cdot[-\delta_{ip}g_{ik}(\delta_{k<m}\delta_{m<i}\delta_{m<l}(1-\delta_{il})\\
&\quad -(\delta_{im}+\delta_{i<m})\delta_{m<k}\delta_{m<l}+\delta_{l<i}\delta_{l<m}(\delta_{kl}+\delta_{k<l})\\
&\quad -(1-\delta_{km})\delta_{l<l}\delta_{l<k}\delta_{l<m})\sum_{|\lambda'''|\varepsilon}V_{lm\lambda'''\varepsilon}\delta_{\lambda+\lambda'+\lambda''+\lambda''',0}\\
&\quad +g_{ip}\delta_{lm}(\delta_{p<l}\delta_{l<i}\delta_{l<k}-(1-\delta_{kp})\delta_{i<p}(\delta_{lp}+\delta_{l<p})\delta_{i<l}\delta_{l<k})\\
&\quad \cdot\sum_{|\lambda'''|}V_{kp\lambda'''\varepsilon}\delta_{\lambda+\lambda'+\lambda''+\lambda''',0}\\
&\quad +g_{pk}\delta_{lm}(\delta_{k<p}(1-\delta_{pi})(\delta_{lp}+\delta_{l<p})\delta_{k<l}\delta_{l<i}-\delta_{p<l}\delta_{l<k}\delta_{l<i})\\
&\quad \cdot\sum_{|\lambda'''|}V_{pi\lambda'''\varepsilon}\delta_{\lambda+\lambda'+\lambda''+\lambda''',0}]\}f\\
&+\sum_{l,i,|\lambda|,|\lambda'|\varepsilon}\frac{\partial^2}{\partial V_{ii\lambda\varepsilon}\partial u^*_{\lambda'}}\{(-h_{\lambda'})\sum_{|\lambda''|}(\delta_{l<i}g_{il}V_{il\lambda''\varepsilon}\\
&\quad -\delta_{i<l}g_{li}V_{li\lambda''\varepsilon})\,\delta_{\lambda+\lambda'+\lambda'',0}\}f\\
&+\sum_{l,i,|\lambda|,|\lambda'|\varepsilon}\frac{\partial^2}{\partial V_{ii\lambda\varepsilon}\partial u_{\lambda'}}\{(-h_{\lambda'})\sum_{|\lambda''|}(\delta_{i<l}g_{il}V_{il\lambda''\varepsilon}\\
&\quad -\delta_{l<i}g_{li}V_{li\lambda''\varepsilon})\,\delta_{\lambda+\lambda'+\lambda'',0}\}f\\
&+\sum_{i,k,|\lambda|,|\lambda'|\varepsilon}\frac{\partial^2}{\partial V_{ki\lambda\varepsilon}\partial u^*_{\lambda'}}\{h_{\lambda'}\sum_{|\lambda''|}g_{ik}\delta_{k<i}(V_{kk\lambda''\varepsilon}-V_{ii\lambda''\varepsilon})\\
&\qquad\qquad \cdot\delta_{\lambda+\lambda'+\lambda'',0}\}f\\
&+\sum_{i,k,|\lambda|,|\lambda'|\varepsilon}\frac{\partial^2}{\partial V_{ki\lambda\varepsilon}\partial u_{\lambda'}}\{h_{\lambda'}\sum_{|\lambda''|}g_{ik}\delta_{i<k}(V_{kk\lambda''\varepsilon}-V_{ii\lambda''\varepsilon})\\
&\qquad\qquad \cdot\delta_{\lambda+\lambda'+\lambda'',0}\}f.
\end{aligned}
$$

Because this equation will be used later as a starting point for further treatments we briefly remind the reader of the notation. The classical amplitude, corresponding to the quantized field amplitudes of mode λ, is u_λ. $V_{ii\lambda\varepsilon}$ is the collective coordinate of the atomic occupation number of level i in the spatial mode λ of the atoms with the level broadening index ε. ε is to be omitted for a homogeneously broadened line. $V_{ik\lambda\varepsilon}$ is the collective off-diagonal element corresponding to the spatial mode λ.

ν_λ is the frequency of the mode λ in the unloaded cavity and κ_λ its decay constant. $h_\lambda g_{lm}$ is defined in Eq. (2.7) and represents the coupling between atoms and field. γ_{ik} is the atomic transition rate from the level i and k (for the definition see [20]), $\hbar\Omega_{k\varepsilon}$ is the energy of the atomic level k with broadening index ε. $n_{\lambda\,\mathrm{th}}$ is the number of thermal photons of mode λ. Inspection of (4.8) reveals that it is an ordinary Fokker-Planck equation containing derivatives up to second order. It describes for sufficiently high numbers of atoms a laser with M modes and N atoms, having each n levels, in a nearly exact fashion. Thus it may serve as a basis for future work on the behaviour of such a laser containing all relevant quantum mechanical information.

Appendix

Explicit Form of the Coefficients M and N

In addition to the Kronecker symbol δ_{ik} we use the symbol $\delta_{i<k}$ which is defined by

$$\delta_{i<k}=\sum_{l=0}^{k-1}\delta_{il}=\begin{cases}1 & \text{if } i<k\\ 0 & \text{if } i\geqq k.\end{cases} \tag{A.1}$$

We further use as previously[4] the abbreviation

$$O^{m\to k}=\delta_{mk}+\sum_{l_1,l_2,\dots l_j} X_{ml_1}\delta_{m<l_1}X_{l_1l_2}\delta_{l_1<l_2}\dots X_{l_jk}\delta_{l_j<k} \tag{A.2}$$

where the sum runs over all possible indices under the restriction $m<l_1<\dots<k$. For descending indices it reads

$$O^{m\leftarrow k}=\delta_{mk}+\sum_{l_1,l_2,\dots l_j} X_{ml_1}\delta_{l_1<m}X_{l_1l_2}\delta_{l_2<l_1}\dots X_{l_jk}\delta_{k<l_j}. \tag{A.3}$$

With this abbreviation the coefficients $M_{ikki,lm}$ and N_{iklm} which had been derived elsewhere[4] can be written in a very compact fashion, namely

$$\begin{aligned}N^{(1)}_{iklm}(\{x\})=&(\delta_{ki}+\delta_{k<i})\,\delta_{il}(\delta_{mk}-\delta_{m<k}X_{km})+\delta_{ik}\,\delta_{im}\,\delta_{m<l}X_{lm}\\&+\{\delta_{l<k}(\delta_{km}-X_{km}\delta_{k<m})\,e^{X_{ll}-X_{kk}}-\delta_{l<m}\,\delta_{m<k}X_{km}\,e^{X_{ll}-X_{mm}}\\&+\sum_n\delta_{l<n}\,\delta_{n<k}\,\delta_{n<m}X_{kn}X_{nm}\,e^{X_{ll}-X_{nn}}\\&+\delta_{i<k}(\delta_{mk}-\delta_{m<k}X_{km})(\delta_{lm}+\delta_{m<l})\}\,O^{l\leftarrow i}\\&-\sum_n\delta_{i<k}(\delta_{mk}-\delta_{m<k}X_{km})\,\delta_{m<n}\,\delta_{n<l}X_{ln}\,O^{n\leftarrow i}.\end{aligned} \tag{A.4}$$

20 See Ref. [10] or [3].

From it $N^{(2)}_{iklm}$ may be found by the following replacements

$$\begin{array}{cccccccc} N^{(1)}_{iklm} & i & k & l & m & \alpha & X_{\alpha\beta} & O^{\alpha\leftarrow\beta} \\ | & | & | & | & | & | & | & | \\ N^{(2)}_{iklm} & k & i & m & l & \tilde{\alpha} & X_{\tilde{\beta}\tilde{\alpha}} & O^{\tilde{\beta}\rightarrow\tilde{\alpha}}. \end{array} \tag{A.5}$$

In a similar manner one finds after lengthy calculations the following explicit form

$$\begin{aligned} M_{ikki,lm}(\{x\}) = (\sum_p O^{k\rightarrow p} O^{p\leftarrow k} e^{X_{pp}}) \{ & \delta_{il}\delta_{lm} e^{-X_{mm}} - [\delta_{il}\delta_{m<i} \\ & + \delta_{im}\delta_{l<i}] e^{-X_{ii}} X_{lm} + [\delta_{m<i}\delta_{i<l} e^{-X_{ii}} + \delta_{l<i}\delta_{i<m} e^{-X_{ii}} \\ & + \delta_{l<m}\delta_{m<i} e^{-X_{mm}} + \delta_{lm}\delta_{l<i} e^{-X_{mm}} \\ & + \delta_{l<i}\delta_{m<l} e^{-X_{ll}}] X_{li} X_{im} - \sum_n \delta_{l<n}\delta_{n<m}\delta_{n<i} \\ & \cdot X_{li} X_{in} X_{nm} e^{-X_{nn}} \\ & - \sum_n \delta_{m<n}\delta_{n<l}\delta_{n<i} X_{ln} X_{ni} X_{im} e^{-X_{nn}} \}, \end{aligned} \tag{A.6}$$

$$\begin{aligned} \sum_p O^{k\rightarrow p} O^{p\leftarrow k} e^{X_{pp}} = \sum_p e^{X_{pp}} (\delta_{kp} + \delta_{k<p} \sum_{\ldots q_i, r_i \ldots} & X_{kq_1}\delta_{k<q_1} \cdots \\ \ldots X_{q_i p}\delta_{q_i<p} X_{pr_1}\delta_{r_1<p} \cdots X_{r_j k}\delta_{k<r_j}) & . \end{aligned} \tag{A.7}$$

The summation runs over all indices so that $k<q_1<\cdots<q_i<p>\gamma_1>\cdots>\gamma_i>k$ is fulfilled.

Expansion of $N^{(1)}_{iklm}$ is a power series of the number of atoms N up to second order. For the diagonal elements $i=k$ we simply obtain the exact result

$$N^{(1)}_{illm}(\{x\}) = \delta_{il}\delta_{im} + (\delta_{im} - \delta_{il})\delta_{m<l} X_{lm}. \tag{A.8}$$

For the corresponding elements $i \neq k$ the result reads

$$\begin{aligned} & N^{(1)}_{iklm}(\{x\}) \\ & = \delta_{il}\delta_{km} + \delta_{il}\delta_{km}\delta_{l<k}(X_{ii} - X_{kk}) \\ & \quad + (\delta_{ik} + \delta_{i<k})\delta_{km}\delta_{i<l} X_{li} - (\delta_{m<k} + \delta_{i<k}\delta_{k<m})\delta_{li} X_{km} \\ & \quad + \delta_{il}\delta_{l<k}\delta_{km} \cdot \tfrac{1}{2}(X_{ll} - X_{kk})^2 + \delta_{l<k}(\delta_{km}\delta_{i<l} X_{li} - \delta_{li}\delta_{k<m} X_{km}) \\ & \quad \cdot (X_{ll} - X_{kk}) - \delta_{il}\delta_{l<m}\delta_{m<k}(X_{ll} - X_{mm}) X_{km} \\ & \quad - \{\delta_{l<k}\delta_{l<m}(1-\delta_{km}) + \delta_{i<k}\delta_{m<k}[\delta_{lm} + (\delta_{im} + \delta_{i<m})\delta_{m<l}]\} \\ & \quad \cdot \delta_{i<l} X_{li} X_{km} + \delta_{li} \sum_p \delta_{l<p}\delta_{p<k}\delta_{p<m} X_{kp} X_{pm} \\ & \quad + \delta_{i<k}\delta_{km} \sum_p (1-\delta_{m<p})\delta_{i<p}\delta_{p<l} X_{lp} X_{pi}. \end{aligned} \tag{A.9}$$

$N^{(2)}_{iklm}$ follows from (A.9) by means of the relation (A.5).

Expansion of M up to terms of second order.
The result reads

$$
\begin{aligned}
M_{ikki,lm}(\{x\}) = {} & \delta_{il}\delta_{lm} + \delta_{il}\delta_{lm}(X_{kk} - X_{ii}) - (\delta_{il}\delta_{m<l} + \delta_{im}\delta_{l<m}) X_{lm} \\
& + \delta_{il}\delta_{lm} \cdot \tfrac{1}{2}(X_{kk} - X_{ii})^2 - (\delta_{il}\delta_{m<l} + \delta_{im}\delta_{l<m}) \\
& \cdot (X_{kk} - X_{ii}) X_{lm} + [\delta_{m<i}\delta_{i<l} + \delta_{l<i}\delta_{i<m} + \delta_{l<m}\delta_{m<i} \\
& + \delta_{lm}\delta_{l<i} + \delta_{m<l}\delta_{l<i}] X_{li} X_{im} + \delta_{il}\delta_{lm} \sum_{p} \delta_{k<p} X_{kp} X_{pk} .
\end{aligned}
\tag{A.10}
$$

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