Quantum Boltzmann-Lorentz model approach to the line-shape problem

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Following our earlier work on quantum kinetic equations and the use of the Boltzmann-Lorentz model to describe the collisional broadening of spectra, we develop a new approach to that problem in which the center-of-mass motion of the emitter as well as the interaction during the collision between the emitter and the buffer gas are treated quantum mechanically. Possible further extensions of our model are discussed.

I. INTRODUCTION

One of the classical problems of atomic and molecular optics, the collisional broadening of spectra, attracts as much attention nowadays as several decades ago.[1] Recent developments in modern spectroscopy call for better understanding of the radiation line shapes of *ultracold atoms*.[2] Indeed, one can envision a situation in which the density of optically active *cold* atoms becomes such that the quantum statistics of them will matter, while the direct interaction between them will still be of negligible importance. Description of such a problem requires that both *center-of-mass* and *internal* atomic degrees of freedom are treated fully quantum mechanically. As a first step in developing such a theory we propose here the quantum generalization of the Boltzmann-Lorentz model of the collisional broadening of spectra.[3]

In the usual approach the change of the atomic spectrum due to interaction between the optically active atom (the emitter) and the buffer gas (the perturbers) is viewed as the interplay of two main phenomena referred to as *velocity modulation* and the *interaction effect*. The velocity modulation concerns the random changes in the velocity of the emitter due to the collisions and the interaction effects arise from the simultaneous perturbation of the pertinent energy levels of the emitter during the collision.

The classical paper on collisional broadening by Rautian and Sobelman[4] contains the analysis of the line-shape problem based on the *classical* linearized Boltzmann equation. The Rautian and Sobelman analysis makes the distinction between (i) the *weak-collision model* and (ii) the *strong-collision model*; the first one is applicable to massive emitters colliding with relatively light buffer-gas particles and the second one pertains to the case in which the emitter is taken to be much lighter than the perturber. The effect of collision is viewed to be so drastic that the velocity of the emitter after the collision is assumed to be completely independent of its velocity before the collision. In a previous publication[3] we have presented a model of the collisional broadening based on the Boltzmann-Lorentz model borrowed from the classical kinetic theory. We were able to treat interaction effects quantum mechanically, while the velocity modulation was considered to be purely classical. Our model was a particular implementation of Berman's[5] idea of a quantum-mechanical transport equation (QMTE). The collisions between emitter and perturbers were treated in Ref. 3 by assuming that the emitter motion was perturbed by random time events with a Poissonian distribution. This was shown to be equivalent to a kinetic theory approach for low perturber density.

In quantum theory we have to use a different approach, and we have found that in order to calculate line shapes it is convenient to work in the Liouville space formalism. This formalism is a widely used tool of quantum many-body theory;[6] it has recently been employed by us for the microscopic evaluation of time-correlation functions[7,8] and the derivation of quantum-kinetic equations.[9]

In this work we shall present a novel formulation of the collisional broadening, based on the Liouville space formalism, which generalizes Berman's QMTE approach. Using the technique developed in Ref. 8 we are able to repeat our analysis[3] but with the center-of-mass motion of the emitter treated fully quantum mechanically.

As in Ref. 3 we assume that the density of the perturbers is low, and therefore we restrict our analysis to the leading order in the perturbers' density. When that density increases one expects several new phenomena, for example, the localization, to become of importance. This has been discussed in Ref. 10 on both the classical[10(b)] and quantum[10(c)] levels. This later reference is of particular importance, for several correlation functions calculated therein have a direct bearing on the line-shape problem.

Our paper is organized similarly as in Ref. 3. We shall begin in Sec. II by presenting our analysis of the velocity modulated line shape in the quantum Boltzmann-Lorentz model. In Sec. III we shall briefly summarize the main result of pure interaction effects from Ref. 3. In Sec. IV we will discuss at some length the combined interaction and velocity modulation effects, again within the full quantum picture. Section V will contain final comments

and conclusions. The necessary technical points are discussed in Appendices A–D.

II. QUANTUM VELOCITY MODULATION

Following Ref. 3 we assume the emitter to be a two-level quantum system (this approximation seems to be particularly well suited for ultracold atoms) which emits a photon when it makes a transition from the upper to the lower energy level on account of the interaction between its dipole moment and the surrounding electromagnetic field. The line shape is given in terms of the dipole-dipole correlation function[6]

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\mathrm{Re}\int_0^\infty dt\, e^{-i\omega t}\langle \mathbf{d}e^{-i\mathbf{k}\cdot\mathbf{r}})\cdot(\mathbf{d}e^{i\mathbf{k}\cdot\mathbf{r}})(t)\rangle \ , \tag{2.1}$$

where $\mathbf{d}$ and $\mathbf{r}$ denote the dipole moment operator and position of the emitter, respectively.

Now, in the Boltzmann-Lorentz model it is assumed that the dilute gas of mutually noninteracting emitters moves in the volume Ω within which an optically inactive buffer gas is present. The buffer-gas atoms (perturbers) are assumed to be much more massive than the emitters and therefore one neglects their motion. Furthermore, the perturbers are randomly distributed throughout the volume and one neglects all effects related to the mutual geometrical arrangements of perturbers. (For instance, the problem of overlapping and nonoverlapping perturber configurations is not being taken into consideration. This plays an important role in the analysis of the localization in the Lorentz model, cf. Ref. 10.) The probability density of the emitter encountering a perturber is then equal to $1/\Omega$. In this section we shall neglect any interaction between the perturber and the emitter during the collision and concentrate on the analysis of the velocity modulation influence on the emitted photon spectrum. The line shape is then obtained from the formula analogous to Eq. (2.1) in which the dipole moment operators are omitted.

In the classical Boltzmann-Lorentz gas theory it is assumed that emitter-perturber encounters are such that the magnitude of the emitter velocity remains unaltered but the velocity direction changes at random.[10] The scattering between emitter and perturber is then the hard-sphere collision. The influence of that type of velocity modulations on the line shape of emitted light was analyzed in Ref. 3 by means of the stochastic matrix approach. Here we shall analyze the velocity modulation assuming that scatterers are quantum particles obeying Fermi-Dirac or Bose-Einstein statistics. We shall neglect, however, any direct interaction between emitters.

In Ref. 3 it was sufficient to analyze effects of the velocity modulation for *one* emitter and to calculate the final result by averaging the final expression over the Maxwell distribution of the emitters. In the quantum case instead we have to analyze the true many-body correlation function which replaces Eq. (2.1), i.e.,

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\mathrm{Re}\int_0^\infty dt\, e^{-i\omega t}\frac{1}{N}\sum_{i,j=1}^{\bar{N}}\langle e^{-i\mathbf{k}\cdot\mathbf{r}_i}(e^{i\mathbf{k}\cdot\mathbf{r}_j})(t)\rangle \ . \tag{2.2}$$

Here the sum runs over all $\bar{N}$ emitters.

The Hamiltonian governing the dynamics of our model is given by

$$\mathcal{H}=\sum_{i=1}^{\bar{N}}\mathcal{H}_i=\mathcal{H}_0+\mathcal{V} \ , \tag{2.3}$$

where $\mathcal{H}_0$ denotes the kinetic energy of the emitters and $\mathcal{V}$ describes the interaction between emitters and perturbers,

$$\mathcal{H}_0=\sum_{i=1}^{\bar{N}}\frac{\mathbf{p}_i^2}{2m} \ , \tag{2.4}$$

$$\mathcal{V}=\sum_{i=1}^{\bar{N}}V(\mathbf{r}_i)=\sum_{i=1}^{\bar{N}}\sum_{j=1}^{N}U(\mathbf{r}_i-\mathbf{R}_j) \ . \tag{2.5}$$

In the above $\mathbf{R}_j$ denotes the position of one of the N perturbers and $U(\mathbf{r}-\mathbf{R})$ describes the interaction between emitter and scatterer. The average in Eq. (2.2) is defined as

$$\langle a\rangle=\mathcal{P}\,\mathrm{tr}(\rho a) \ , \tag{2.6}$$

where $\mathcal{P}$ denotes the average over the randomly distributed perturbers,

$$\mathcal{P}=\mathcal{P}^{1,\ldots,N}=\mathcal{P}^1\cdots\mathcal{P}^N=\prod_{i=1}^{N}\frac{1}{\Omega}\int_\Omega d\mathbf{R}_i \ , \tag{2.7}$$

and tr denotes the trace over a complete set of emitter states. The operator ρ is the canonical density matrix

$$\rho=Z^{-1}\exp(-\beta\mathcal{H})$$

with the partition function $Z(\{\mathbf{R}_i\})=\mathrm{tr}\exp(-\beta\mathcal{H})$ depending on the perturber positions. The latter property does not play an important role in our analysis since Z is self-averaging.[11] The time evolution of any operator a is given by

$$a(t)=e^{i\mathcal{L}t}a(0) \ , \tag{2.8}$$

where $\mathcal{L}$ denotes the Liouville (super) operator defined as

$$\mathcal{L}a\equiv\frac{1}{\hbar}[\mathcal{H},a] \ . \tag{2.9}$$

The fact that the Hamiltonian Eq. (2.3) is a sum of single-particle contributions allows us to prove the following useful identity (cf. Appendix A):

$$\mathrm{tr}\rho\sum_{i=1}^{\bar{N}}a_i\sum_{j=1}^{\bar{N}}b_j=\mathrm{tr}_1[f_1a_1(1+\eta f_1)b_1]+[\mathrm{tr}_1(f_1a_1)][\mathrm{tr}_1(f_1b_1)] \ , \tag{2.10}$$

where a_i and b_i are one-particle operators and tr_1 means the one-particle trace. Furthermore,

$$f_1=\frac{1}{\exp[\beta(\mathcal{H}_1-\mu)]-\eta} \tag{2.11}$$

is the one-particle Fermi-Dirac or Bose-Einstein distribution operator and μ denotes the chemical potential. The coefficient η equals ∓1 for fermion and boson emitters, respectively. Note that $\mathcal{H}_1$ still contains the interaction

between emitter 1 and the perturbers.

With the aid of identity (2.10) we can write the integrand of the line-shape formula as

$$\sum_{i,j}\langle e^{-i\mathbf{k}\cdot\mathbf{r}_i}(e^{i\mathbf{k}\cdot\mathbf{r}_j})(t)\rangle=\mathcal{P}\,\mathrm{tr}_1[f_1 e^{-i\mathbf{k}\cdot\mathbf{r}_1}(1+\eta f_1)(e^{i\mathbf{k}\cdot\mathbf{r}_1})(t)] + |\mathcal{P}\,\mathrm{tr}_1(f_1 e^{-i\mathbf{k}\cdot\mathbf{r}_1})|^2 . \tag{2.12}$$

The last term on the right-hand side of this equation is proportional to $\delta_{\mathbf{k},0}$ and can be omitted since we are only interested in the case $\mathbf{k}\neq 0$. The pure modulation line shape is then given by the formula

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\mathrm{Re}\int_0^\infty dt\, e^{-i\omega t}\mathcal{P}\,\mathrm{tr}[fe^{-i\mathbf{k}\cdot\mathbf{r}}(1+\eta f)(e^{i\mathbf{k}\cdot\mathbf{r}})(t)] , \tag{2.13}$$

where we have dropped the index 1.

Equation (2.13) is the quantum generalization of the classical velocity modulated emission line shape [cf. Eq. (2.2) of Ref. 3]. We can rewrite Eq. (2.13) in a form which is better suited for further analysis by using the identity below, which holds for any analytic function of the Hamiltonian $\mathcal{H}$:

$$e^{-i\mathbf{k}\cdot\mathbf{r}}g(\mathcal{H}(\mathbf{p}))e^{i\mathbf{k}\cdot\mathbf{r}}\equiv g(\mathcal{H}(\mathbf{p}+\hbar\mathbf{k})) . \tag{2.14}$$

With the aid of the "shifted" Hamiltonian $\mathcal{H}(\mathbf{p}+\hbar\mathbf{k})$ we can define generalized Liouville operators:

$$\mathcal{L}^{\mathbf{k}}a=\frac{1}{\hbar}[\mathcal{H}(\mathbf{p}+\hbar\mathbf{k})a-a\mathcal{H}(\mathbf{p})] ,$$
$$\mathcal{L}_0^{\mathbf{k}}a=\frac{1}{\hbar}[\mathcal{H}_0(\mathbf{p}+\hbar\mathbf{k})a-a\mathcal{H}_0(\mathbf{p})] , \tag{2.15}$$
$$\mathcal{L}_V a=\frac{1}{\hbar}[V(\mathbf{r}),a] .$$

Equation (2.13) can now be rewritten as

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\lim_{\epsilon\to 0+}\mathrm{Re}\mathcal{P}\,\mathrm{tr}\left[f(1+\eta f^{\mathbf{k}})\frac{1}{\epsilon+i\omega-i\mathcal{L}^{\mathbf{k}}}\right] , \tag{2.16}$$

where $f^{\mathbf{k}}=f(\mathcal{H}(\mathbf{p}+\hbar\mathbf{k}))$. Note that in Eq. (2.16) $(\epsilon+i\omega-i\mathcal{L}^{\mathbf{k}})^{-1}$ acts on the unit operator $\hat{1}$.

The further analysis of expression (2.16) is based on the formalism developed in Refs. 7 and 8. One evaluates the trace in Eq. (2.16) by using emitter momentum eigenstates $|\mathbf{q}\rangle$ and diagonal and off-diagonal projection operators

$$\langle\mathbf{q}|P^e a|\mathbf{q}'\rangle=\langle\mathbf{q}|a|\mathbf{q}'\rangle\delta_{\mathbf{q},\mathbf{q}'} ,$$
$$Q^e=1-P^e . \tag{2.17}$$

The memory-function-like form of the line shape reads then $(z=i\omega+\epsilon)$

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\lim_{\epsilon\to 0+}\mathrm{Re}\mathcal{P}\,\mathrm{tr}S(\mathbf{k})[1+N(\mathbf{k},z)]\left[\frac{1}{z-D^e(\mathbf{k},z)}\right] , \tag{2.18}$$

where the memory kernel $D^e(\mathbf{k},z)$ does not depend explicitly on perturber coordinates while the amplitude correction $N(\mathbf{k},z)$ does. This, and the appearance of the $\mathbf{k}$-dependent static structure factor $S(\mathbf{k})$ are important modifications as compared to the classical theory. Explicitly we have

$$D^e(\mathbf{k},z)=P^e i\mathcal{L}_0^{\mathbf{k}}P^e+\mathcal{P}P^e\left[\frac{z}{z-i\mathcal{L}^{\mathbf{k}}Q^e}\right]i\mathcal{L}_V P^e . \tag{2.19}$$

$$N(\mathbf{k},z)=Q^e\left[\frac{1}{z-i\mathcal{L}^{\mathbf{k}}Q^e}\right]i\mathcal{L}_V P^e , \tag{2.20}$$

$$S(\mathbf{k})=f(1+\eta f^{\mathbf{k}}) . \tag{2.21}$$

The derivation of formula (2.18), which is the main result of this section, is outlined in Appendix B.

We can now discuss our result in various interesting limits. First we shall analyze the large-$|\mathbf{k}|$ limit. Since $\mathcal{H}(\mathbf{p}+\hbar\mathbf{k})\propto|\mathbf{k}|^2$ for large $|\mathbf{k}|$, $S(\mathbf{k})\to f(\mathcal{H})$. Furthermore, using the results of Appendix B, we find that

$$D(\mathbf{k},z)\to P^e i\mathcal{L}_0^{\mathbf{k}}+O(1/k^2)$$

while $N(\mathbf{k},z)\to O(1/k^2)$. Thus the large-$k$ limit of the line-shape function equals

$$I(\mathbf{k},\omega)\underset{k\to\infty}{\sim}\frac{1}{\pi}\lim_{\epsilon\to 0+}\mathrm{Re}\sum_{\mathbf{q}}[\mathcal{P}f(H)]_{\mathbf{qq}}\frac{1}{\epsilon+i\omega-i(\hbar/m)\mathbf{q}\cdot\mathbf{k}-i(\hbar/2m)\mathbf{k}^2} . \tag{2.22}$$

In the classical regime, i.e., $\hbar\to 0$, replacing $\mathcal{H}$ by $\mathcal{H}_0$ in f, we recover from (2.22), the line-shape profile from Ref. 3.

On physical grounds we are more interested in the opposite limit, that is, long wavelengths and low frequencies. We shall analyze our velocity-modulated line shape in the limit of low perturber density $(n=N/\Omega\to 0)$ and $k,\omega\to 0$. For small n

$$S(\mathbf{k})\to f(\mathcal{H}_0)[1+\eta f(\mathcal{H}_0(\mathbf{p}+\hbar\mathbf{k}))]$$

and in the long-wavelength approximation we can use

$$S(\mathbf{k})\approx S_0(\mathbf{k}=0)=-(1/\beta)[\partial f(\mathcal{H}_0)/\partial\mathcal{H}_0] .$$

The line shape then becomes [with $P^e N(\mathbf{k},z)=0$, since $P^e S_0(\mathbf{k})=S_0(\mathbf{k})P^e$ and $P^e Q^e=0$]

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\lim_{\epsilon\to 0+}\mathrm{Re}\,\mathrm{tr}S_0(\mathbf{k})\frac{1}{z-D^e(\mathbf{k},z)}[1+O(n)] , \tag{2.23}$$

where the small-n limit of the memory function $D^e(\mathbf{k},z)$ reads

$$D^e(\mathbf{k},z)=P^e i\mathcal{L}_0^{\mathbf{k}}+n\int d\mathbf{R}_1 P^e i\mathcal{L}_1\frac{z}{z-iQ^e\mathcal{L}(1)}P^e +O(n^2,kn)\ , \tag{2.24}$$

where $\mathcal{L}(1)=\mathcal{L}_0+\mathcal{L}_1$. In the thermodynamic limit Eq. (2.24) simplifies considerably, since the projector Q^e can be dropped. We can then express our line shape in terms of the Liouville space T matrix

$$T_1(z)=-i\mathcal{L}_1\frac{1}{z-\mathcal{L}(1)}(z-\mathcal{L}_0)\ . \tag{2.25}$$

The memory function $D^e(\mathbf{k},z)$ becomes then (with $\mathcal{L}_0P^e=0$)

$$D^e(\mathbf{k},z)=P^e i\mathcal{L}_0^{\mathbf{k}}-n\int d\mathbf{R}_1 P^e T_1(z)P^e+O(n^2,kn)\ . \tag{2.26}$$

The simple form of this formula is in fact a bit misleading. Appendix C contains the necessary algebra leading to the final expression for the quantum-mechanical velocity-modulated line shape, which can be written in a closed form provided the scattering length approximation for $T_1(z)$ is used. We obtain

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\mathrm{Re}\Omega\int d\mathbf{p}\frac{1}{(2\pi)^3}[S^0(\mathbf{k})]_{\mathrm{pp}}\frac{\tau^1(\mathbf{p})}{1-\gamma\overline{\tau^1(\mathbf{p})}}\ , \tag{2.27}$$

where

$$[S_0(\mathbf{k})]_{\mathrm{pp}}=(e^{\beta[\varepsilon(\mathbf{p})-\mu)}-\eta)^{-1}\times\left[1+\frac{\eta}{\exp[\beta\varepsilon(\mathbf{p}+\mathbf{k})-\mu]-\eta}\right]\underset{k\to 0}{\sim}-\frac{1}{\beta}\frac{\partial f(\varepsilon)}{\partial\varepsilon} \tag{2.28}$$

is the static form factor, $\varepsilon(\mathbf{p})=\hbar^2\mathbf{p}^2/2m$, and

$$\tau^1(\mathbf{p})=\frac{1}{i(\omega-\hbar\mathbf{p}\cdot\mathbf{k}/m-\hbar\mathbf{k}^2/2m)+\gamma(p)}\ , \tag{2.29}$$

$$\overline{\tau^1(p)}=\frac{m}{\hbar kp}\arctan\left[\frac{\hbar kp/m}{\gamma(p)+i(\omega-\hbar k^2/2m)}\right]\ . \tag{2.30}$$

Expression (2.30) is obtained by averaging $\tau^1(\mathbf{p})$ over the directions of $\mathbf{p}$. The coefficient $\gamma(p)$ is expressed in terms of the s-wave scattering length a for the potential U of the emitter-perturber interaction (cf. Appendix C),

$$\gamma(p)=4\pi na^2(p\hbar/m)\equiv 4\pi na^2 v(p)\ , \tag{2.31}$$

where $v(p)$ is the emitter velocity. Note the well-known[12,13] factor 4 between the quantum-mechanical value of γ obtained here and the classical hard-sphere expression used in Ref. 3.

The above scattering length approximation result for the quantum-velocity modulated line shape can now be compared with the classical Boltzmann-Lorentz model predictions.[3] If the density of the emitters is low, that is, when the mean distance between emitters is larger than the thermal de Broglie wavelength of the emitters $\bar{n}\lambda_{\mathrm{dB}}^3\ll 1$, we can approximate the distribution function $f(\varepsilon)$ as

$$f(\varepsilon)\approx\exp(-\beta\varepsilon+\beta\mu)=\bar{n}(h^2/2\pi mk_BT)^{3/2}\exp(-\beta\varepsilon)\ ,$$

where we have used the classical expression for the chemical potential μ. Using this we obtain the nondegenerate form of the line shape

$$I^{\mathrm{ND}}(\mathbf{k},\omega)=4\bar{N}\left[\frac{2\pi k_BT}{m}\right]^{-3/2}\times\mathrm{Re}\int_0^\infty dv\, v^2 e^{-mv^2/2k_BT}\frac{\overline{\tau^1(v)}}{1-\gamma(v)\overline{\tau^1(v)}}\ . \tag{2.32}$$

Here $\overline{\tau^1(v)}$ and $\gamma(v)$ are obtained from the corresponding expressions (2.30) and (2.31) by replacing $\mathbf{p}$ by the velocity $\mathbf{v}=\hbar\mathbf{p}/m$.

Following Ref. 3 we can analyze Eq. (2.32) in two extreme limits of the parameter $\zeta=na^2/k$. For small ζ, when the perturbers have negligible influence on the emitters, the observed line shape is Gaussian with the photon recoil taken into account. Indeed the center of the line is shifted by $\hbar k^2/2m$ but the width remains as in the classical Doppler broadening

$$\Gamma_G=(8\pi k_BT\ln 2/m)^{1/2}k\ ,$$

$$I^{\mathrm{ND}}(k,\omega)=\frac{1}{\pi k}\left[\frac{\pi m}{2k_BT}\right]^{1/2}\times\exp\left[-\frac{m}{2k_BTk^2}\left[\omega-\frac{\hbar k^2}{2m}\right]^2\right]\ . \tag{2.33}$$

Note that this result can also be obtained from the large-k limit, Eq. (2.22), of the exact expression for the spectral function.

In the more interesting limit when $\zeta\gg 1$, i.e., when the radiation wavelength is larger than the mean distance between the perturbers, the collisions should have a predominant effect on the line shape. Indeed, as in the classical case, we now obtain the observed line-shape profile which is Lorentzian with the width $\Gamma_L=3\pi\Gamma_G\sqrt{\pi\ln 2}/\zeta$,

$$I^{\mathrm{ND}}(k,\omega)=\frac{(1/2\pi)\Gamma_L}{(\omega-\hbar k^2/2m)^2+\frac{1}{4}\Gamma_L^2}\ . \tag{2.34}$$

As in the small-ζ limit the photon recoil is taken into account.

It follows from the above discussion that in the case of pure velocity modulation, for a low concentration of perturbers and in the nondegenerate limit, the quantum expression for the observed emission line shape is nearly the same as in the classical theory. The width of the line does not change, but the photon recoil is properly taken into account. For a typical visible light frequency and an emitter mass of the order of a few hydrogen atom masses

the photon recoil shift is of the order of 10^6–10^7 Hz, clearly within the experimentally accessible range. We shall now follow Ref. 3 and discuss the pure interaction effects.

III. INTERACTION EFFECTS

In this section we shall briefly recall the analysis of the pure interaction effects.[3] Thus we shall neglect the influence of the velocity changes at the collisions but discuss the effects of the interaction between the emitter and the perturber during the collision.

We model the emitter as a quantum two-level system, which can be represented in terms of the pseudospin S^z ($S=\frac{1}{2}$) eigenstates $|\pm\rangle$ such that its unperturbed Hamiltonian reads

$$\mathcal{H}_0=\hbar\bar{\omega}S^z , \tag{3.1}$$

where $\bar{\omega}$ is the frequency of the emitted radiation. In a collision process, when the emitter approaches the perturber, an interaction takes place which may induce a transition between the energy levels of the emitter—a (pseudo)spin-flip process. Obviously, the detailed description of that process requires a careful quantum-mechanical analysis of the atomic collision process. Here we take the point of view that perturbers are essentially classical objects and that during the collision they produce an effective field $\mathbf{H}_j$ which couples to the emitter. A schematic view of those processes is shown in Fig. 1.

Following Refs. 3 and 6 we therefore write the Hamiltonian for the emitter as[14]

$$\mathcal{H}(t)=\mathcal{H}_0+\sum_{\alpha}\hbar(1+\mathbf{H}_1\cdot\mathbf{S})\delta(t-t_\alpha) , \tag{3.2}$$

where the sum runs over time instants, labeled by α, at which collisions take place. Similarly, as in Refs. 3 and 10, those time instants are assumed to be Poisson distributed.

The Heisenberg representation of the dipole operator $\mathbf{d}$ for the emitter reads

$$\mathbf{d}(t)=\mathcal{U}(t)\mathbf{d}(0) , \tag{3.3}$$

where $\mathcal{U}(t)$ is the evolution operator given as

$$\mathcal{U}(t)=\mathcal{T}\exp\left[i\int_0^t\mathcal{L}_s(t')dt'\right] . \tag{3.4}$$

Here $\mathcal{L}_s(t)$ is the Liouville operator corresponding to the Hamiltonian Eq. (3.2) and $\mathcal{T}$ denotes the time-ordering operator. The line shape is now obtained from the spectral function $\phi(z)$

$$\phi(z)=\int_0^\infty dt\, e^{-zt}\langle\mathbf{d}(0)\cdot\mathbf{d}(t)\rangle , \tag{3.5}$$

where the angular brackets denote averaging over the quantum states of the emitter and over the statistics of the collisions.

In this section we will be interested in the emission line shape. Since the dipole operator for a two-level atom can be conveniently expressed in terms of the (pseudo) spin raising and lowering operators, the spectral function can then be written as

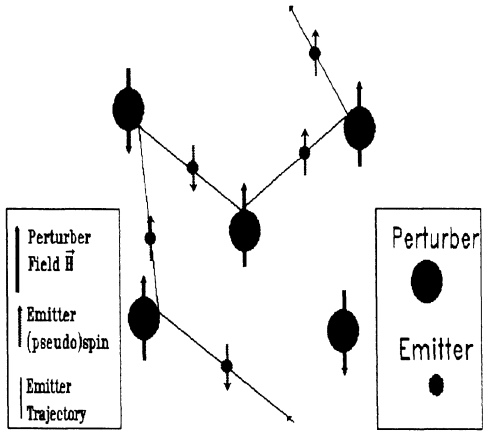

FIG. 1. The Boltzmann-Lorentz model. Large solid circles denote the perturber gas atoms while small circles denote emitter particles. Heavy arrows denote instantaneous direction of the effective field **H** and smaller arrows indicate the spins of the emitters.

$$\phi_{+-}(z)=\mathrm{tr}\rho\int_0^\infty e^{-zt}\langle+|S^+|-\rangle\langle-|[\mathcal{U}(t)S^-]|+\rangle . \tag{3.6}$$

In the above ρ is the density matrix and we have explicitly displayed the spin states arrangement involved in the process of averaging.

In Ref. 3 we have argued that in the high-temperature limit, that is, when $\hbar\bar{\omega}\ll k_BT$, the line shape can be written as

$$I(\omega)=\mathrm{Re}\int_0^\infty f_M(v)[i(\omega+\bar{\omega})+\tfrac{1}{3}\gamma(v)H^2]^{-1}dv , \tag{3.7}$$

where f_M denotes the Maxwell distribution function of the center-of-mass velocity of the emitter, $\gamma(v)$ is the same as in Sec. II, and H^2 is the mean-square value of the effective fields $\mathbf{H}_j$. The integral in Eq. (3.7) can be evaluated exactly leading to the line shape given in terms of the exponential integral

$$E_1(\{(\omega+\bar{\omega})/[\tfrac{1}{3}\gamma(v_{\mathrm{th}})H^2]\}^2) .$$

This shape is very similar to the Lorentzian line shape with the width proportional to $\gamma(v_{\mathrm{th}})H^2$, where v_{th} is the mean thermal emitter velocity, as shown in Fig. 2 where both shapes are plotted as functions of their corresponding distance $\Delta\omega$ from the line center.

We shall now propose an extension of the above analysis which will take into account, among other things, the quantum degeneracy of the center-of-mass motion of the emitters. We found that in order to do so it is more convenient to use the approach developed in Ref. 7, and used in Sec. II, rather than just the outlined random kick approach.

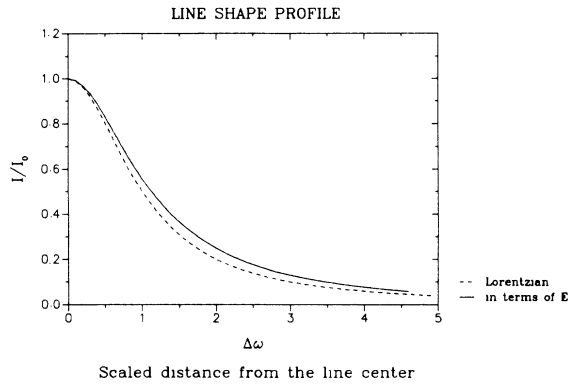

FIG. 2. Comparison of the Lorentzian and exponential integral line shape discussed in Sec. III with the Lorentzian line shape. $\Delta\omega$ is the corresponding distance from the line center and both profiles are normalized. $E_1(x)$ is defined as in Ref. 21.

IV. COMBINATION OF VELOCITY MODULATION AND INTERACTION EFFECTS

In this section we shall describe the combined effect of the velocity modulation and interaction on the line shape of the emitter. To begin with, we write the Hamiltonian for our model as

$$\mathcal{H}=\sum_{i=1}^{\bar{N}}[\mathcal{H}_0(i)+\mathcal{V}(i)]\ , \tag{4.1}$$

where $\mathcal{H}_0(i)$ is the free ith emitter Hamiltonian and $\mathcal{V}(i)$ is the interaction Hamiltonian.

$$\mathcal{H}_0(i)=\frac{\mathbf{p}^2}{2m}+\hbar\bar{\omega}S^z\ , \tag{4.2}$$

$$\mathcal{V}(i)=\sum_{j=1}^{N}W_j(i)\equiv\sum_{j=1}^{N}U(\mathbf{r}_i-\mathbf{R}_j)(1+\mathbf{H}_j\cdot\mathbf{S}_i)\ . \tag{4.3}$$

As usual, the canonical density matrix is $\rho=\exp(-\beta\mathcal{H})/Z$, with the Hamiltonian given by Eq. (4.1). Following the usual assumption as to the distribution of the fields $\mathbf{H}_j$, we generalize the projection operator (2.7) and write

$$\mathcal{P}=\mathcal{P}^{1,\ldots,N}=\mathcal{P}^1\cdots\mathcal{P}^N=\prod_{j=1}^{N}\left[\frac{1}{4\pi}\int d\mathbf{\Gamma}_j\frac{1}{\Omega}\int d\mathbf{R}_j\right]\ , \tag{4.4}$$

where the first integration is over the random directions of the perturber fields $\mathbf{H}_j$. ($\mathbf{\Gamma}_j$ is the unit vector.)

Now, the general definition of the line shape given by Eq. (2.1) together with Eq. (2.6) and the identity Eq. (2.10) allow us to write the line-shape function as

$$I(\mathbf{k},\omega)=\frac{1}{\pi}\mathrm{Re}\phi(\mathbf{k},\omega)\ , \tag{4.5}$$

with the spectral function $\phi(\mathbf{k},\omega)$

$$\phi(\mathbf{k},\omega)=\int_0^\infty dt\, e^{-i\omega t}\mathrm{Tr}\mathcal{P}[f\,\mathbf{d}e^{-i\mathbf{k}\cdot\mathbf{r}}(1+\eta f)(\mathbf{d}e^{i\mathbf{k}\cdot\mathbf{r}})(t)]\ , \tag{4.6}$$

where, as previously,

$$f(1)=1/\exp\{\beta[\mathcal{H}(1)-\mu]-\eta\}$$

and now $\mathrm{Tr}\equiv\mathrm{tr}_e\mathrm{tr}_s$ denotes the trace with respect to center-of-mass and internal (pseudospin) degrees of freedom of the emitter. Note that now the dipole operator (representable in terms of the spin raising and lowering operators) does not commute with $f(1)$.

The emission line shape is obtained by extracting from Eq. (4.5) the piece of the spectral function ϕ_{+-} and then taking its real part. Following the notation of Sec. II, Eqs. (2.14)–(2.16), we write

$$\phi_{+-}(\mathbf{k},\omega)=\lim_{\epsilon\to0+}\mathrm{Tr}\mathcal{P}\left[fS^+(1+\eta f^{\mathbf{k}})\frac{1}{z-iL^{\mathbf{k}}}S^-\right]\ , \tag{4.7}$$

and that should be compared with the expression for the pure velocity modulation line shape Eq. (2.16).

Now, exactly the same procedure leading from Eq. (2.26) to Eq. (2.28) (see also Appendix B) can be applied to expression (4.7) [with P defined in Eq. (4.4)]. The result is

$$\phi_{+-}(\mathbf{k},\omega)=\lim_{\epsilon\to0+}\mathrm{Tr}\mathcal{P}\bar{S}(\mathbf{k},\bar{\omega})\times[1+N(\mathbf{k},z)]\left[\frac{1}{z-D(\mathbf{k},z)}\right]S^-\ , \tag{4.8}$$

where $N(\mathbf{k},z)$ and $D(\mathbf{k},z)$ are defined as in Eqs. (2.19) and (2.20) but with the Liouvillean determined by the Hamiltonian Eq. (4.1). $\bar{S}(\mathbf{k},\bar{\omega})$ is the spin-dependent "static structure factor" equal to

$$\bar{S}(\mathbf{k},\bar{\omega})=fS^+(1+\eta f^{\mathbf{k}})\ . \tag{4.9}$$

Note that $\bar{\omega}$, the unperturbed emitter level splitting, should not be confused with the frequency ω.

Similarly, as in Sec. II, the memory function D is already averaged over the random variables pertaining to the perturbers, while $N(\mathbf{k},\omega)$ still depends on positions and effective fields of the perturbers. The spectral function given by Eq. (4.8) is exact. We shall now discuss approximate expressions for the line shape which follow from it. Again, as in Sec. II, we assume the low density of the perturbers. We can then approximate $\bar{S}(\mathbf{k},\bar{\omega})$ from Eq. (4.8) by its low-density limit

$$\bar{S}(\mathbf{k},\bar{\omega})\to\bar{S}_0(\mathbf{k},\bar{\omega})=f_0S^+(1+\eta f_0^{\mathbf{k}})\ .$$

It follows then that

$$\langle\sigma|\bar{S}_0(\mathbf{k},\bar{\omega})|\sigma'\rangle=\hbar\delta_{\sigma,+}\delta_{\sigma',-}(e^{\beta(H_0^e+\hbar\bar{\omega}/2-\mu)}-\eta)^{-1}[1+\eta(e^{\beta(H_0^e(\mathbf{p}+\hbar\mathbf{k})-\hbar\bar{\omega}/2-\mu)}-\eta)^{-1}]$$
$$=\delta_{\sigma,+}\delta_{\sigma',-}S_0(\mathbf{k},\bar{\omega}) , \tag{4.10}$$

where $\mathcal{H}_0^e=\mathbf{p}^2/2m$. Note that for high temperatures ($k_BT\gg\hbar\bar{\omega}$) $S_0(\mathbf{k},\bar{\omega})\to S_0(\mathbf{k})$, where $S_0(\mathbf{k})$ is the same as in Sec. II.

The spectral function for the emission line shape then becomes

$$\phi_{+-}(\mathbf{k},\omega)=\lim_{\epsilon\to 0+}\mathrm{tr}_e S_0(\mathbf{k},\bar{\omega})\left\langle -\left|\left|\left[\frac{1}{z-D(\mathbf{k},z)}S^-\right]\right|\right|+\right\rangle[1+O(n)] . \tag{4.11}$$

with the memory function given by a formula analogous to Eq. (2.19). Note, however that the free Liouvillean of the emitter consists now of two parts related to the center-of-mass and pseudospin degrees of freedom, and the interaction part of the Liouvillean is given by

$$\mathcal{L}_V=\sum_{j=1}^{N}\mathcal{L}_j \tag{4.12}$$

with $\mathcal{L}_j A=[W_j, A]$, where $W_j(1)$ is given by Eq. (4.3).

In our further analysis we proceed as in Sec. II. For details see Appendix D. The final result of our calculations is then the following expression for the spectral function ϕ_{+-}:

$$\phi_{+-}(\mathbf{k},\omega)=\sum_{\mathbf{p}}[S_0(\mathbf{k},\bar{\omega})]_{\mathbf{p},\mathbf{p}}\times\frac{\tau^1(\mathbf{p})}{1-\gamma(p)[1-(\hbar^2/12)H^2]\overline{\tau^1(p)}} , \tag{4.13}$$

where

$$\tau^1(\mathbf{p})=\left[i\left[-\frac{\hbar}{m}\mathbf{k}\cdot\mathbf{p}-\frac{\hbar^2}{2m}k^2+\omega+\bar{\omega}\right]+\gamma(p)\left[1+\frac{\hbar^2}{4}H^2\right]\right]^{-1} \tag{4.14}$$

and (integration is carried out over the unit sphere in momentum space)

$$\overline{\tau^1(p)}=\frac{1}{4\pi}\int d\Omega(\hat{p})\tau^1(\mathbf{p})=\frac{m}{\hbar kp}\arctan\left[\frac{\hbar kp/m}{\gamma(p)(1+\hbar^2H^2/4)+i(\omega+\bar{\omega}-\hbar k^2/2m)}\right] . \tag{4.15}$$

If we neglect the $\mathbf{k}$ dependence of the static structure factor, i.e., replace $S_0(\mathbf{k},\bar{\omega})$ by $S_0(0,\bar{\omega})\equiv S_0(\bar{\omega})$, we obtain from Eq. (4.5) and (4.13), replacing sums by integrals, the following expression for the line shape:

$$I(\mathbf{k},\omega)=\frac{\Omega}{2\pi^3}\mathrm{Re}\int_0^\infty dp\,p^2[S_0(\bar{\omega})]_{p,p}\times\frac{\overline{\tau^1(p)}}{1-\gamma(p)[1-(\hbar^2/12)H^2]\overline{\tau^1(p)}} . \tag{4.16}$$

Equation (4.16) is the quantum generalization of Eq. (4.14) from Ref. 3, the Voigt-like profile.[15] In the nondegenerate limit, replacing the structure factor by the Maxwell distribution, as in Sec. II, we obtain from Eq. (4.18) the line shape as in Ref. 3, but with the photon recoil properly taken into account.

V. CONCLUSIONS

We have derived the emission line shape for the quantum generalized Boltzmann-Lorentz model. We have used the quantum Liouville space formulation proposed in Refs. 7 and 8, but the rest of our analysis was analogous to Ref. 3. In the nondegenerate limit we have obtained line shapes analogous to those in the classical Boltzmann-Lorentz theory, however with the photon recoil corrections properly included.

The Boltzmann-Lorentz model for the line shape was recently reformulated[16] using the stochastic dynamics approach. That approach is completely equivalent to the analysis of Ref. 3 and there is no obvious way how to include the quantum center of mass motion into that formulation.

Our current work uses extensively the techniques of the quantum-kinetic theory, and we have obtained our results in the leading order with respect to the perturbers' density. The generalization to higher densities is tedious and requires utmost care. At the moment we see no real need to go to the higher order in density expansion in applications to the line-shape problems.

We can see several possible applications of our theory. We have already mentioned in Sec. I that in recent experiments on ultracold atoms the quantum corrections due to the center-of-mass motion of the cold atoms might be important. Although we have presented here calculations of the line shape only, it should be fairly clear that we can repeat the whole analysis for other correlation functions. In particular, one should be able to calculate the current-current correlation function and therefore obtain the quantum expression for the diffusion and spin-diffusion coefficients. The classical theory of those coefficients, for the Boltzmann-Lorentz model, was recently discussed in Ref. 17. A further possible application is to consider those correlation functions for strong quantum degeneracy. That should be of interest for the physics of the polarized hydrogen gas.[18]

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APPENDIX A

This appendix contains the proof of the identity Eq. (2.10). We use the standard second quantization notation to replace an operator $\sum_i a_i$ as follows:

$$\sum_{i=1}^{\bar{N}} a_i \rightarrow \sum_{k,l} \alpha^\dagger a_{kl} \alpha_l \ , \tag{A1}$$

where $a_{kl}=\langle k|a_1|l\rangle$ are the matrix elements of the one-particle operator a_1 with respect to the complete set of one-particle Hamiltonian eigenstates

$$\mathcal{H}_1|k\rangle = E_k|k\rangle \ . \tag{A2}$$

$\alpha_k^\dagger$ and α_l are the creation and annihilation operators fulfilling the (anti)commutation relations

$$\begin{aligned} \alpha_k \alpha_l^\dagger &= \eta \alpha_l^\dagger \alpha_k + \delta_{kl} \ , \\ \alpha_k \alpha_l &= \eta \alpha_l \alpha_k \ , \end{aligned} \tag{A3}$$

with $\eta=-1$ for fermions and $+1$ for bosons. The occupation number of the state $|k\rangle$ will be denoted by $n_k=\alpha_k^\dagger \alpha_k$. The system Hamiltonian can then be written as $\mathcal{H}=\sum_k n_k E_k$. Now, the trace in Eq. (2.10) is understood as being taken in the grand canonical ensemble with the chemical potential μ. Thus

$$\mathrm{tr}\rho \sum_{i=1}^{\bar{N}} a_i \sum_{j=1}^{\bar{N}} b_j = \frac{1}{Z} \mathrm{tr} \left[\exp\left[\beta \sum_k n_k(\mu - E_k)\right] \sum_{k,l} \alpha_k^\dagger a_{kl} \alpha_l \sum_{mn} \alpha_m^\dagger b_{mn} \alpha_n \right] \ , \tag{A4}$$

where Z is the grand canonical partition function. The trace in Eq. (A4) is taken over all possible sets of occupation numbers $\{n_k\}$. It turns out that in the fourfold sum in (A4) the only nonvanishing terms are those with (i) $k=l$, $m=n$, or (ii) $k=n$, $l=m$, $k\neq l$. [The term $k=1$ is already included in (i).] In view of (A3) one can rewrite (A4) as

$$\frac{1}{Z} \mathrm{tr} \exp\left[\beta \sum_k n_k(\mu - E_k)\right] \left[\sum_k n_k a_{kk} \sum_m n_m b_{mm} + \sum_{k\neq m} a_{km} b_{mk} \eta n_k n_m \right] . \tag{A5}$$

Denoting the thermal average in the grand canonical ensemble by a bar and recalling that

$$\bar{n}_k = f_k \equiv 1/\{\exp[\beta(E_k-\mu)]-\eta\}$$

and $\overline{n_k^2}=f_k+(1+\eta)f_k^2$ we can rewrite (A5) in the form

$$\sum_{k,m} [f_k a_{kk} f_m b_{mm} + a_{km} b_{mk} f_k (1+\eta f_m)] \ . \tag{A6}$$

The sum in (A6) can now be written as trace tr_1 over one-particle states, and that immediately leads to Eq. (2.10). Another proof of (2.10) is possible directly in the canonical ensemble, without second quantization, by using the formalism developed in Ref. 8.

APPENDIX B

In this appendix we derive formula (2.18) starting from the expression

$$\phi^e \equiv \mathcal{P}P^e S \frac{1}{z-i\mathcal{L}} P^e, \quad z=\epsilon+i\omega \ , \tag{B1}$$

which occurs on the right-hand side of Eq. (2.16). The static factor S is defined in Eq. (2.21) and for notational simplicity the k dependence of S and $\mathcal{L}$ is suppressed in the following. Making use of the operator identity

$$\frac{1}{X-Y} = \left[1+\frac{1}{X-Y}Y\right]\frac{1}{X} \tag{B2}$$

in (B1), we find

$$\phi^e = \frac{1}{z} \mathcal{P}P^e S \left[1+\frac{1}{z-i\mathcal{L}} i\mathcal{L}_0 + \frac{1}{z-i\mathcal{L}} i\mathcal{L}_V\right] P^e \ . \tag{B3}$$

Next we replace the resolvent of the third term in (B3) by the identity

$$\frac{1}{z-i\mathcal{L}} = \left[Q^e + \frac{z}{z-i\mathcal{L}} P^e\right] \frac{1}{z-i\mathcal{L}Q^e} \ , \tag{B4}$$

which follows from the relation $P^e+Q^e=1$ and (B2) with $X\equiv z-i\mathcal{L}Q^e$ and $Y\equiv i\mathcal{L}P^e$. We are then left with

$$\phi^e = PP^eS \frac{1}{z-i\mathcal{L}} \left[\frac{1}{z} i\mathcal{L}_0 + P^e \frac{1}{z-i\mathcal{L}Q^e} i\mathcal{L}_V\right] P^e + \frac{1}{z} PP^eS \left[1+Q^e \frac{1}{z-i\mathcal{L}Q^e} i\mathcal{L}_V\right] P^e \ . \tag{B5}$$

The central point in our derivation is now that in the thermodynamic limit ($N,\Omega\rightarrow\infty$ with $n=N/\Omega$ finite) we can replace the first set of large parentheses in (B5) by $P(\cdots)$. This leads to a closed equation for ϕ^e and subsequently to the desired result Eq. (2.18). We shall proceed here in a way analogous to Secs. III and IV of Ref. 7.

To begin with we define superoperators F and G as

$$F(\mathbf{r}-\mathbf{R}_1,\ldots,\mathbf{r}-\mathbf{R}_N) = S \frac{1}{z-i\mathcal{L}} \ , \tag{B6}$$

$$G(\mathbf{r}-\mathbf{R}_1,\ldots,\mathbf{r}-\mathbf{R}_N) = \frac{1}{z-i\mathcal{L}Q^e} i\mathcal{L}_V^e \ , \tag{B7}$$

and analyze the expression PFP^eG occurring within the first set of large parentheses. Density expansions of both F and G give

$$PFP^eG=P\sum_{s=1}^{N\to\infty}\sum_{k=0}^{s}\sum_{d=0}^{N\to\infty}N^dF_{k+d}(\mathbf{r}-\mathbf{R}_1,\ldots,\mathbf{r}-\mathbf{R}_k,\mathbf{r}-\mathbf{R}_{s+1},\ldots,\mathbf{r}-\mathbf{R}_{s+d})P^eN^sG_s(\mathbf{r}-\mathbf{R}_1,\ldots,\mathbf{r}-R_s)\ , \tag{B8}$$

where the superoperators F_l and G_s depend only on the relative coordinates between the emitter ($\mathbf{r}$) and scattering centers $\{\mathbf{R}_l\}$. The explicit forms of F_l and G_s are of no importance here (although they can be easily evaluated by means of cluster expansions, cf. Ref. 8). What matters is that F_l ($l\geq 1$) and G_s are essentially different from zero only when $|\mathbf{r}-\mathbf{R}_i|\leq a$, $\forall i$, where a is the interaction range of the potential $U(\mathbf{r}-\mathbf{R}_l)$. F_0 is obviously independent of U.

We now consider a typical term occurring in (B8),

$$N^{s+d}PF_{k+d}P^eG_s=n^{d+s}\int d\mathbf{R}_1\cdots d\mathbf{R}_{s+d}F_{k+d}P^eG_s\ , \tag{B9}$$

with $0\leq k\leq s$, and estimate its volume dependence for different k's. For this it is most convenient to use the phase-space representation[19] (for details see Sec. 2 of Ref. 7)

$$y^{\rm ph}(\mathbf{r},\mathbf{p})=\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}\langle\mathbf{p}+\mathbf{k}/2|y|\mathbf{p}-\mathbf{k}/2\rangle\ , \tag{B10}$$

$$S^{\rm ph}y^{\rm ph}(\mathbf{r},\mathbf{p})=(Sy)^{\rm ph}(\mathbf{r},\mathbf{p})\ , \tag{B11}$$

where y is an ordinary operator and S a superoperator. In that representation the projection operator P^e assumes the simple form $(P^e)^{\rm ph}=(1/\Omega)\int d\mathbf{r}$ which immediately allows us to determine the volume dependence of (B9):

$$n^{d+s}\int d\mathbf{R}_1\cdots d\mathbf{R}_{s+d}F^{\rm ph}_{k+d}(P^e)^{\rm ph}G^{\rm ph}_s=\begin{cases}O(1) & \text{for } k=0\\ O\left[\dfrac{1}{\Omega}\right] & \text{for } k>0\ .\end{cases} \tag{B12}$$

It follows that in the thermodynamic limit the only term contributing to (B8) is that with $k=0$. Thus

$$PFP^eG=PFPP^eG+O\left[\frac{1}{\Omega}\right]\ . \tag{B13}$$

Using this in expression (B5), recalling that $[\mathcal{L}_0^{\mathbf{k}},P^e]=0$ and dropping terms of the order Ω^{-1}, we find

$$\phi^e=\phi^eP\left[\frac{1}{z}i\mathcal{L}_0+P^e\frac{1}{z-i\mathcal{L}Q^e}i\mathcal{L}_V\right]P^e\ ,$$

$$+\frac{1}{z}PP^eS\left[1+Q^e\frac{1}{z-i\mathcal{L}Q^e}i\mathcal{L}_V\right]P^e\ . \tag{B14}$$

From the above Eq. (2.18) follows immediately.

APPENDIX C

We shall outline here the derivation of Eq. (2.28), starting from expression (2.26) for the function $D^e(\mathbf{k},z)$. The strategy here is to express the Liouville T matrix defined in Eq. (2.25) in terms of the ordinary t matrix given by

$$t_1(z;\mathbf{R}_1)=U(\mathbf{r}-\mathbf{R}_1)\frac{1}{\mathcal{H}_0+U(\mathbf{r}-\mathbf{R}_1)-z}(\mathcal{H}_0-z)\ , \tag{C1}$$

and then to evaluate it in a scattering length expansion. Thereby we shall retain only the leading order in the scattering length a. In a second step, we determine the inverse of $z-D^e(\mathbf{k},z)$ needed in Eq. (2.23).

As shown in Eq. (A.6) of Ref. 7 one has

$$\lim_{\epsilon\to 0+}[P^eT_1(\varepsilon)P^ey]_{\rm pp}=\frac{2\pi}{\hbar}\sum_{\mathbf{p}_1}|t_1^\dagger(\varepsilon(\mathbf{p}))_{\mathbf{pp}_1}|^2\times\delta(\varepsilon(\mathbf{p})-\varepsilon(\mathbf{p}_1))\times[y(\mathbf{p})-y(\mathbf{p}_1)]\ , \tag{C2}$$

where $t_1^\dagger(E)=\lim_{\epsilon\to 0}t_1(E+i\epsilon;\mathbf{R}_1)$. y is an ordinary operator and $y(\mathbf{p})=\langle\mathbf{p}|y|\mathbf{p}\rangle=y_{\rm pp}$.

Now, neglecting the ω dependence of $D^e(\mathbf{k},z=\epsilon+i\omega)$, we obtain from (2.26) and (C2)

$$\lim_{\epsilon\to 0+}[D^e(\mathbf{k},\epsilon)y]_{\rm pp}=\left[i\frac{\hbar}{m}\mathbf{p}\cdot\mathbf{k}+i\frac{\hbar}{2m}k^2\right]y_{\rm pp}-\frac{2\pi\Omega}{\hbar}n\sum_{\mathbf{p}_1}|t^\dagger(\varepsilon(\mathbf{p}))_{\mathbf{pp}_1}|^2\delta(\varepsilon(\mathbf{p})-\varepsilon(\mathbf{p}_1))[y(\mathbf{p})-y(\mathbf{p}_1)]\ , \tag{C3}$$

where we have used the fact that

$$\int d\mathbf{R}_1|t_1^\dagger(E)_{\mathbf{pp}'}|^2=\Omega|t^\dagger(E)_{\mathbf{pp}'}|^2 \tag{C4}$$

with $t^\dagger(E)=\lim_{\epsilon\to 0+}t_1(E+i\epsilon,\mathbf{R}_1=0)$.

Next we perform the scattering length expansion[12,20] of $t^\dagger(\varepsilon(\mathbf{p}))_{\mathbf{pp}_1}$ which is valid for a strong purely repulsive interaction potential (e.g., hard-sphere interaction). In lowest order in the scattering length a (s-wave approximation) one has $t^\dagger(\varepsilon(\mathbf{p}))_{\mathbf{pp}_1}=2\pi\hbar^2a/m\Omega+O(a^2)$. Using that and replacing sums over momenta by integrals, $\Sigma_{\mathbf{p}}\to[\Omega/(2\pi)^3]\int d\mathbf{p}$, we obtain, after some algebra,

$$i\omega-\lim_{\epsilon\to 0+}D^e(\mathbf{k},z)=\Lambda^1-\Lambda^2\ , \tag{C5}$$

with

$$(\Lambda^1y)_{\rm pp}=\left[i\left[\omega-\frac{\hbar}{m}\mathbf{p}\cdot\mathbf{k}-\frac{\hbar}{2m}k^2\right]+\gamma(\mathbf{p})\right]y(\mathbf{p})\ , \tag{C6}$$

$$(\Lambda^2y)_{\rm pp}=\gamma(\mathbf{p})\overline{y(p)}\ . \tag{C7}$$

$\gamma(\mathbf{p})$ is given in Eq. (2.31) and $\overline{y(p)}$ denotes the average of

$y(\mathbf{p})$ over the directions of $\mathbf{p}$.

We need now to invert expression (C4) occurring in Eq. (2.23). We have

$$I(\mathbf{k},\omega)=\mathrm{Re}\sum_{pp_1} S_0(\mathbf{k})_{pp}\left[\frac{1}{\Lambda^1}\right]_{pp|pp}\left[\frac{1}{1-\Lambda^2\frac{1}{\Lambda^1}}\right]_{pp|p_1p_1}, \tag{C8}$$

where the tetradic matrix of a superoperator S is defined by

$$(Sy)_{kk'}=\sum_{pp'} S_{kk'|pp'}y_{pp'} . \tag{C9}$$

Using expressions (C6) and (C7) we obtain

$$\left[\frac{1}{\Lambda^1}\right]_{pp|pp}=\frac{1}{i[\omega-(\hbar/m)\mathbf{k}\cdot\mathbf{p}-(\hbar/2m)k^2]+\gamma(p)} \equiv\tau^1(p) , \tag{C10}$$

$$\sum_{p_1}\left[\frac{1}{1-\Lambda^2\frac{1}{\Lambda^1}}\right]_{pp|p_1p_1}=\sum_{p_1}\sum_{n=0}^{\infty}\left[\left[\Lambda^2\frac{1}{\Lambda^1}\right]^n\right]_{pp|p_1p_1} =\sum_{n=0}^{\infty}[\gamma(p)\overline{\tau^1(p)}]^n =\frac{1}{1-\gamma(p)\overline{\tau^1(p)}} . \tag{C11}$$

On substituting (C10) and (C11) into (C8) and choosing $\mathbf{k}$ parallel to the z axis we obtain Eqs. (2.27)–(2.30).

APPENDIX D

In this appendix we derive Eq. (4.13) starting from Eq. (4.11). First we expand

$$D(\mathbf{k},z)=P^e\left[i\mathcal{L}_0^{\mathbf{k}}+\mathcal{P}i\mathcal{L}_V\frac{z}{z-iQ^e\mathcal{L}^{\mathbf{k}}}\right]P^e , \tag{D1}$$

to the lowest order in $n,\omega,\bar{\omega},k$, scattering length a, and effective field $\mathbf{H}$. Then we determine the inverse of $z-D(\mathbf{K},\omega)$ needed in Eq. (4.11).

In a first step we find

$$D(\mathbf{k},z)=iP^e\mathcal{L}_0^{\mathbf{k}}-NP^1P^eT_1(\varepsilon)P^e[1+O(n,\omega,\bar{\omega},k)] , \tag{D2}$$

where the Liouville T matrix is defined in Eq. (2.25) but now with $\mathcal{L}_1$ given in Eq. (4.12). $P^eT_1P^e$ shall now be expressed in terms of ordinary t matrices. Since P^e projects on diagonal matrix elements only in the momentum but not in (pseudo)spin eigenstates, we cannot use the simple relation (C2), but have to start from the more general expression derived in Ref. 7 [Eq. (A2) of that reference]. Thus instead of (C2) we obtain

$$\lim_{\epsilon\to 0+}[P^eT_1(\varepsilon)P^ey]_{pp}=-\lim_{\epsilon\to 0+}\left[it_1(\varepsilon(\mathbf{p})^-)_{pp}y(\mathbf{p})-iy(\mathbf{p})t_1(\varepsilon(\mathbf{p})^+)_{pp} +2\pi\sum_{p_1}t_1(\varepsilon(\mathbf{p})^-)_{pp_1}y(\mathbf{p}_1)t_1(\varepsilon(\mathbf{p})^+)_{p_1p}\delta(\varepsilon(\mathbf{p})-\varepsilon(\mathbf{p}_1))\right] , \tag{D3}$$

where $t_1(E^{\pm})$ equals

$$t_1(E^{\pm})=W_1\frac{1}{\mathcal{H}_0^e+W_1-E^{\pm}}(\mathcal{H}_0^e-E^{\pm}),\quad E^{\pm}=E\pm i\frac{\epsilon}{2} . \tag{D4}$$

Here $\mathcal{H}_0^e$ denotes the free Hamiltonian of Eq. (2.4). From now on the superscript e refers to external emitter degrees of freedom. Note that expression (D3) is still an operator in the (pseudo)spin space, thus, e.g., $y(\mathbf{p})$ and $t_1(E^{\pm})_{pp_1}$ do not commute in general.

Now, we will expand t_1 in powers of the effective field $\mathbf{H}$ and the scattering length a. Thereby we also make use of the generalized optical theorem

$$t_1^e(E^-)-t_1^e(E^+)=t_1^e(E^-)\frac{2i\epsilon/2}{(\epsilon/2)^2+(\mathcal{H}_0^e-E)^2}t_1^e(E^+) , \tag{D5}$$

and of the relations $P^1\mathbf{H}\cdot\mathbf{S}=0$, $\mathcal{L}_0^s y=\hbar\bar{\omega}[S^z,y]$, and $(S^x)^2=(S^y)^2=(S^z)^2=\frac{1}{4}$. After some lengthy but straightforward algebra we obtain (retaining only leading terms in H and a) an expression which replaces (C5), with operators $\Lambda^{1,2}$ now having the following form:

$$(\Lambda^1y)_{pp}=\left\{i\left[\omega-\mathcal{L}_0^s-\frac{\hbar}{m}\mathbf{k}\cdot\mathbf{p}-\frac{\hbar}{2m}k^2\right] +\gamma(\mathbf{p})\left[1+\left[\frac{\hbar}{2}\right]^2H^2\right]\right\}y(\mathbf{p})(\Lambda^2y)_{pp} =\gamma(\mathbf{p})\left[\overline{y(p)}+\tfrac{1}{3}H^2\sum_{i=1}^{3}S^i\overline{y(p)}S^i\right] . \tag{D6}$$

Finally, we have to determine the inverse of the operator $i\omega-\lim_{\epsilon\to 0+}D(\mathbf{k},z)$ occurring in Eq. (4.11). Proceeding as in Appendix C and making use of the relation $\sum_{i=1}^{3}S^iS^{\pm}S^i=-S^{\pm}/4$, we arrive at the desired result (4.13).

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