Phase space density limitation in laser cooling without spontaneous emission

Thierry Chanélière*, Daniel Comparat* and Hans Lignier*
Laboratoire Aimé Cotton, CNRS, Univ. Paris-Sud,
ENS Paris Saclay, Université Paris-Saclay,
Bât. 505, 91405 Orsay, France
*These authors contributed equally to this work
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We study the possibility to enhance the phase space density of non-interacting particles submitted to a classical laser field without spontaneous emission. We clearly state that, when no spontaneous emission is present, a quantum description of the atomic motion is more reliable than semi-classical description which can lead to large errors especially if no care is taken to smooth structures smaller than the Heisenberg uncertainty principle. Whatever the definition of position - momentum phase space density, its gain is severely bounded especially when started from a thermal sample. More precisely, the maximum phase space density, can only be improved by a factor \( M \) for \( M \)-level atoms. This bound comes from a transfer between the external and internal degrees of freedom. To circumvent this limit, one can use non-coherent light fields, informational feedback cooling schemes, involve collectives states between fields and atoms, or allow a single spontaneous emission event.

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It is usually believed that the phase space density (PSD) of non-interacting particles cannot be increased by using only pure Hamiltonian evolution and any PSD increase would require a dissipative mechanism \([1,2]\). In the context of laser cooling, this dissipation is usually ensured by spontaneous emission. Nevertheless, in recent years, several papers showed experimentally optical cooling without spontaneous emission. This counter intuitive results was also supported by theoretical arguments and semi-classical simulations using classical laser fields. The perspective of cooling very different species including molecules has actively stimulated the discussions \([3-7]\).

In this letter, we specifically address the issue of cooling non-interacting particles without spontaneous emission submitted to classical laser fields (i.e. equivalent to quantum fields in coherent mode \([8,10]\)). We first determine the evolution of a particle distribution in phase space. In particular, we show that a quantum treatment of external degrees of freedom is more reliable than a classical treatment that may lead to erroneous predictions. A quantum description of position and momentum requires to revisit the definition of the classical PSD, to define quantum analogs of PSD and to discuss various characterizations. In any case, we prove that the gain in PSD of an initial thermal distribution is possible but clearly limited to the number \( M \) of internal levels.

First of all, it is important to recall that the evolution of non-interacting particles can be derived from a single particle statistics. In this framework, we neglect the single realizations of many-particle evolution that may cause PSD modification because of ergodicity, Zermelo-Poincaré recurrence or Fluctuation theorems \([11]\) as through coarse grained PSD \([12,13]\) or by PSD volume surrounding particles (such as ellipsoid emittance growth in beams) \([14]\). Therefore, we assume the ensemble evolution as entirely derived from the one-particle density matrix \( \rho \) in the quantum case and, in the classical case, from the (statistical averaged single particle) classical PSD \( \rho(r,v,t) \).

The most general evolution of the classical PSD undergoing a (non-random) external force \( F(r,v,t) \) is given by the continuity equation:

\[
\frac{D\rho}{Dt} = \frac{\partial \rho}{\partial t} + (v \cdot \frac{\partial}{\partial r}) \rho + \frac{F}{m} \cdot \frac{\partial \rho}{\partial v} = -\rho \frac{\partial}{\partial v} \frac{F}{m} \tag{1}\]

where \( \frac{D\rho}{Dt} \) is the material derivative. This clearly shows that a velocity-dependent force is necessary to change \( \rho \). The Doppler cooling scheme, using for example the classical Lorentz oscillator model, is a textbook example of velocity-dependent force. However, in Hamiltonian mechanics, according to the (Vlasov-)Liouville’s theorem \( \frac{D\rho}{Dt} = 0 \) for non-interacting particles, \( \rho \) is constant. This is consistent with the continuity equation because friction forces cannot be described in Hamiltonian mechanics \([17]\). Since quantum mechanics is also based on a Hamiltonian description, one may wonder how a change of PSD could be explained. A major difference actually comes from the treatment of internal degrees of freedom that cannot be rigorous in classical physics. Regarding the electromagnetic interactions, the time evolution of the internal degrees of freedom is generally calculated by the quantum master equation acting on the density matrix because it may include also non-unitary evolutions due to spontaneous emission. The semi-classical evolution of the external degrees of freedom is then usually obtained by Ehrenfest’s theorem. This framework provides satisfying predictions for Doppler cooling where the semi-classical PSD change is essentially attributed to spontaneous emission. However even without spontaneous emission, several semi-classical studies and propositions suggest that the PSD can be modified (π-pulse, rapid adiabatic passage (RAP), Stimulated RAP, bi-chromatic
FIG. 1: Left: Pulse sequence, a $\pi$-pulse coming from the left followed by a $\pi$-pulse coming from the right. Right: basic idea of PSD increase. The first $\pi$-pulse transfer one atom from $|p, g\rangle$ to $|p, e\rangle$ without modifying the atom already in state $|p, g\rangle$, so increasing the total population (PSD) in $|p\rangle$ by a factor 2. Trying to add a third atom in the same momentum $|p\rangle$ cell, by applying a second $\pi$-pulse counter-propagating, simply swaps states with no gain in $|p\rangle$ population. $\omega_{rec} = \frac{\hbar k^2}{2m}$ is the recoil frequency.

3461516). Their common idea is that a coherent force, resulting from absorption and stimulated emission, depends on the particle velocity via the Doppler effect. So a large increase of PSD seems possible from the continuity equation (1). In the following, we will show that the concept of semi-classical force is only partly correct and that the Ehrenfest’s theorem can lead to an important overestimation of the cooling efficiency. We will show that a proper quantum mechanical treatment exhibit a limited gain in PSD, its maximum being the number $M$ of internal levels.

The basic physical mechanism and maximum gain of PSD can be understood using an ensemble of non-interacting two-level atoms (with ground $|g\rangle$ and excited $|e\rangle$ internal states) and momentum states $|p\rangle$. Because, the atoms do not interact with each other and do not undergo spontaneous emission, the one particle Hamiltonian where the fields are classical is sufficient the describe the dynamics. This latter can be found in the Supplemental Material [17, Eq.(2)].

In Fig.[1] we sketch an example of population increase in the external state: a light pulse (with Doppler detuning and Rabi frequency $\Omega$ wisely adjusted to address a narrow line recoil transition) may bring two atoms in the same momentum state $|p\rangle$ while the internal state of the displaced atom is changed. Any attempt to increase further the population of $|p\rangle$ is vain: populations are swapped between states, because the rates of absorption and stimulated emission are equal. This roughly explains the limitation in PSD gain of a factor 2 for 2-level atoms.

We now confirm this limit by accurate calculations for two pulses in one dimension as depicted in Fig.[1]. The classical evolution and the quantum evolution of an initial two-dimensional (thermal) Gaussian initial distribution in $(r, p)$ are given in Fig.[2]. The quantum evolution is based on the density matrix master equation $\dot{\rho}(p, t)$ [17, Eq.(12)] and the Wigner function $W(r, p, t)$ [17, Eqs.(13-15)]. We determine that the maximum Wigner PSD gain reaches 2.5. The semi-classical evolution uses of Newton’s equation of motion with a force [17, Eq.(21)] resulting from the Ehrenfest’s theorem and Bloch equations [17, Eq.(20)] using the $\hbar k \to 0$ limit of the Wigner quantum evolution (see [17]). The evolution of the semi-classical PSD distribution is calculated with a billion of test particles. The final plot corresponds to the number of atoms in a position-momentum cell whose size has been arbitrary chosen as $1/(5k)$ in position and $\hbar k/10$ in momentum (a) and smoothed distribution (b) as well as the total (ground plus excited states) Wigner (c) and Husimi (d) functions are shown after a pair of $\pi -$ pulses (left-right) with Rabi frequency $2\omega_{rec}$ and pulses detuning $-2\omega_{rec}$.
slower external motion; so the Wigner function evolution reduces to the semi-classical one as demonstrated in [17]. On the contrary, without spontaneous emission, correlations may appear between internal and external variables [19] invalidating the semi-classical approach. We also ran similar simulations on bichromatic and adiabatic transfer schemes; it also appears that the classical and quantum evolutions differ significantly.

The physical relevance of the previous calculations should now be discussed in the light of the position-momentum uncertainty principle because both the quantum and semi-classical distributions exhibit structures smaller than the minimum uncertainty. This problem is often present in the distributions processed in cooling (brightening) studies [3–7]. This issue can be solved by convoluting the PSD distribution with a Gaussian function (Weierstrass transform) corresponding to the Heisenberg limit $\sigma_\sigma \sigma_\rho = \hbar/2$, which gives the smoothed coarse grained distributions also shown in Fig.2(b,d), where we choose $k\sigma = \sigma/\hbar = 1/\sqrt{2}$. Applied to Wigner function, we obtain the so-called $Q(r,p,t)$ Husimi distribution which is the optimum probability distribution for joint measurement of position and momentum [20]. The effect is quite striking since the classical $\rho$ and quantum $Q$ approaches, although different, are now maximally bounded by a gain of 2. Although these smoothed distributions are similar in the particular case of our toy model, they could be much more different for other protocols. Even with a smoothing post-procedure, the semi-classical evolution should fail at the time when particles initially in the ground state and contained in an Heisenberg-bounded PSD region become subject to different forces (or Rabi frequencies).

A reliable characterization of the quantum PSD, more precisely a proper definition of the PSD gain, is necessary to avoid misinterpretations and controversies. To do so, we first consider the entropy $S$ (per particles and per unit of $kg$) because it can quantitatively describe the PSD. For instance, this quantity is denoted $D$ in the Boltzmann’s formula

$$S = -\ln D$$ \hspace{1cm} (2)

Alternatively, the Sackur-Tetrode formula $S = -\ln D + \frac{5}{2}$ gives the thermal classical PSD used by the ultracold community (the number of particles contained in a cube of side equal to the de Broglie wavelength) equals to unity when quantum degeneracy is reached. In this work, we will use Eq. (2) to calculate $D$ from the different definitions of $S$ that we examine below. We first consider the Von Neuman entropy $S_{VN} = -\text{Tr} [\hat{\rho} \ln(\hat{\rho})] = -\sum_i |r_i| \ln |r_i|$ where $r_i$ are the eigenvalues of the single particle density matrix $\hat{\rho}$. These eigenstates generally do not correspond to physical observables $|E_i>$ as the energy eigenstates for example. So other quantities are commonly used, such as the informational Shanon entropy $S_{Sh} = -\sum_i p_i \ln p_i$ where $p_i = \langle E_i | \hat{\rho} | E_i \rangle$ is the population of the $i^{th}$ eigenstate. Consequently, we define $D_{VN}$ and $D_{Sh}$ from Eq. (2). These particular cases belong to two distinct and general categories: eigenvalue-based (or spectral) entropy and population-based (or informational) entropy. The first kind is independent of the representation basis and thus invariant under Hamiltonian evolution while the second kind depends on the representation and consequently is likely to change over time. In these conditions, one can wonder if a quantum entropy can decrease or not. To answer this question, we reconsider the evolution of the quantum PSD distributions in Fig.2. However, in order to calculate $D_{Sh}$ and $D_{VN}$ more easily, we now assume initially the atoms fully delocalized in position, which implies that the initial density matrix is Gaussian diagonal when expressed in $|p>$ basis. We check that this small modification has almost no effect on the evolution of the gain (Fig.2 shows that the smoothed spatial distribution is almost not affected by the time evolution). As expected, we see Fig.3(a) that the Von Neuman entropy is invariant while the Shanon entropy is not. More fundamentally, an initial thermal state provides the largest possible PSD and prohibits further PSD increase [11]. Indeed, the minimum Shanon entropy is achieved by a thermal Gaussian state [21] and it then equals the Von-Neuman entropy. So in our case $D_{Sh}(t) \leq D_{Sh}(0) = D_{VN}(0)$. Yet it is noticeable that $D_{Sh}$ can increase as observed locally in Fig.3(a) between $\omega_{rec}t = \pi/4$ and $\omega_{rec}t = \pi/2$ when the density matrix is no more Gaussian diagonal. Cooling is indeed possible on non-thermal states (as the one produced at time $\omega_{rec}t = \pi/4$).

Finally, we would like to discuss the decrease of $D_{Sh}$ and the invariance of $D_{VN}$, which seems to contradict the results of Fig.2 where all the distribution maxima in-
crease. This apparent contradiction comes from the fact that the whole density matrix we considered is composed of two subspaces: the full atomic system $AB$ ($\hat{\rho} = \hat{\rho}_{AB}$) is formed by the external degrees of freedom part $A$ and the internal degrees of freedom $B$ of size (rank) $M$ (here $M = 2$). As the PSD distributions in Fig. 8 are functions of the coordinates $(r, p)$ linked to $A$, it is thus more appropriate to evaluate $S^{(A)}$ (or $D^{(A)}$), i.e. $S$ (or $D$) restricted to $A$ by using $\hat{\rho}_A = T_{B\rightarrow D} \hat{\rho}$ instead of $\hat{\rho}$. The quantity $S^{(A)}$ is not submitted to the constraints imposed to $S$ because entropy can be exchanged between the two subspaces. For instance, $S_{VN}$ verifies the subadditivity and the Araki-Leib inequality $S^{(AB)}_{VN} - S^{(B)}_{VN} \leq S^{(A)}_{VN} \leq S^{(AB)}_{VN} + S^{(B)}_{VN}$ where the maximum of $S^{(B)}_{VN} = \log M$ [22, 24]. Using Eq. (2), we thus find the fundamental inequality

$$\frac{1}{M} D^{(AB)} \leq D^{(A)} \leq MD^{(AB)}$$

that bounds the evolution. The possible modification of $S^{(A)}_{VN}$ is obviously linked to the mutual entropy $S^{(A)}_{VN} + S^{(B)}_{VN}$ - $S^{(AB)}_{VN}$ defining the maximal cooling (work) that can be achieved in quantum thermodynamics [21]. The triangle inequality [21] indicates that a subtly correlated system could even lead to an increase of $D^{(A)}$ by a factor $M^2$ [25]. However, under the canonical conditions where only one internal state is populated, the gain of $D^{(A)}$ is bounded to $M$ since $S^{(AB)}_{VN}(0) = S^{(A)}_{VN}(0)$ and $S^{(AB)}_{VN}(t) = S^{(B)}_{VN}(0)$. This is consistent with the results shown in Fig. 3(b) where the gain on $D^{(VN)}_{VN}$ is greater than one but lower than $M = 2$.

The gain limit of $M$ is a fundamental result of our study. This latter also holds for $S^{(A)}_{Sh}$ and consequently $D^{(A)}_{Sh}$ can only increase by a factor $M$ for an initial thermal state because $D^{(A)}_{Sh} \leq D^{(A)}_{VN}$ both quantities being equal for an initial diagonal (or thermal) state. This argument is general and can be extended to other entropy definitions or functions such as linear Rényi, min-entropy, state purity or spectral radius of the state. Indeed the key argument is that all the defined entropy (Hartley, Tsallis, Wehrl, Manfredi-Feix, Rényi, Shannon, Gibbs, Von Neumann, min, max, linear, ...) [26, 27] are concave functions of the parameters (power or logarithm). As a consequence, Jensen’s inequality and Schur-Horn’s theorem impose that an informational entropy is larger than the corresponding spectral entropy [21, 24, 28]. Similarly, for the maximal population of $\hat{\rho}_A$ [2], we have $\max |\hat{\rho}_A(t)| \leq M \max |\hat{\rho}_A(0)|$ which is demonstrated in [28]. In a same manner, it can be shown that for position-momentum coherent states $|\alpha(r, p)\rangle$, the evolution of the Husimi function $Q$ as well as the Wehrl entropy (that is the classical limit $h \rightarrow 0$ of the Von Neumann quantum entropy [29]) are bounded by the same factor $M$. [72]. This is consistent with our numerical results in Fig. 3 showing the evolution of the quantities $\max |\hat{\rho}_A|$, $\max |Q^{(A)}|$, $S^{(A)}_{VN}$ and $S^{(A)}_{Sh}$ where, to precise the notations we use the reduced density matrix such as $\max |\hat{\rho}_A| = \max_p \langle p, g | \hat{\rho}(t) | p, g \rangle + \langle p, e | \hat{\rho}(t) | p, e \rangle$. Similarly, $S_{Sh} = - \sum_p \langle p, g | \hat{\rho}(p, g) | \hat{\rho}(p, g) \rangle - \sum_p \langle p, e | \hat{\rho}(p, e) | \hat{\rho}(p, e) \rangle$ and for the Husimi function, we plot the maximum of $Q(r, p, t) = Q^{(g)}(r, p, t) + Q^{(e)}(r, p, t) = \frac{1}{2} \sum_{\alpha, e} |\langle \alpha, g | \hat{\rho}(\alpha, e) + \langle e, \alpha | \hat{\rho}(\alpha, e) | \hat{\rho}(\alpha, e) \rangle|$, where $|\alpha(r, p) = \frac{\alpha}{\alpha} + i \frac{\beta}{\alpha}$ is a coherent state. As an important precaution, we mention that using pseudo phase space density definitions, as based on filtering of some specific states (such as for the ground state only $S^{(g)}_{Sh} = - \sum_p \langle p, g | \hat{\rho}(p, g) | \hat{\rho}(p, g) \rangle$, it is possible to find larger increase than a factor 2 [2].

In conclusion, in absence of spontaneous emission and using classical laser fields, we have shown that a quantum description is more reliable than a semi-classical description of the atomic motion which can lead to large errors. We have also shown that the total eigenvalues-based PSD (Von-Neuman, Rényi, min, purity, spectral radius for example) can not increase. This conclusion can be extended to informational population-based PSD (max $|\rho|$, Sh entropy or max $|Q|$) when the initial state is a diagonal state. Still, a sample initially prepared in a thermal state and thereby without quantum correlation can exhibit a gain of the position-momentum PSD up to the number $M$ of internal states. The direct and fundamental consequence of this analysis, holding for any kind of free particles or in a time-dependent trapping potential, for instance cooling mechanisms based on coherent transfer of photon momenta without spontaneous emission (such as adiabatic passages, bichromatic, $\pi$-pulses [31, 32, 33, 34] or cavity cooling [22, 45, 49, 49–52]), have a limited efficiency and could lead only to a PSD gain of $M$ (or ultimately $M^2$ if initial correlations exists in the initial state). An obvious way to overcome this limit is to allow a single spontaneous emission event per particle [31, 33] because the third ancilla spontaneous emission space has almost an infinite dimension to extract entropy (see [34, 35]). A second option for cooling is to create entanglement between atoms and light field [15, 16] or by using non statistical methods such as informational cooling (stochastic cooling being one famous example) [17, 18] or cavity cooling [22, 45, 49, 49–52]. A final alternative would be to use non-classical quantum fields. Because absorption or stimulated emission rate are not equivalent anymore (Fock states for example), the last step sketched in Fig. 3 now allows to put more atoms at the same phase space location [10]. In other words, when the optical field is no longer considered as a parameter, the total system is now composed of 3 sub-systems (external degree of freedom, internal degree of freedom and quantized field). Our previous demonstrations could then be applied: the (external) PSD can be increased by the number of available microstates in the other (internal and field) spaces. If these
latter are sufficiently large, there is a priori no theoretical limit on cooling even without spontaneous emission [3, 4, 22, 43, 49, 51, 53].

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NON-RELATIVISTIC HAMILTONIAN OF NON-INTERACTING PARTICLES

We here recall the equations of motion for laser cooling of atoms. The reader can refer to textbooks such as [54].

Quantized or (semi-)classical hamiltonian

We here study the quantum Hamiltonian \( \hat{H} \) of a two generic levels \(|1\rangle\) and \(|2\rangle\) (representing the ground \(|g\rangle\) and the excited \(|e\rangle\) states in [55]) of a particle (mass \( m \)) under the effect of electromagnetic fields. The generalisation to \( M \) level system is straightforward but will not be detailed for the sake of simplicity. We separate the "motional" (or trapping) fields that do not couple \(|1\rangle\) and \(|2\rangle\), such as trapping potential \( V_1, V_2 \) produced for example by magnetic coils, magnets or electrodes through Zeeman \((-\mu_B B)\) or Stark effect \((-dE)\), and the laser fields \( \vec{E} \) that do couple \(|1\rangle\) and \(|2\rangle\).

For \( N \) non-interacting particles the full hamiltonian can be written as

\[
\hat{H} = \sum_{i=1}^{N} \hat{H}^{(i)} + \hat{H}_{\text{field}} + \sum_{i=1}^{N} \hat{H}_{\text{int-field}}^{(i)},
\]

where \( \hat{H}^{(i)} \) is the hamiltonian \( \frac{\vec{p}_i^2}{2m} + V_1(\vec{r}_i, t)|1\rangle\langle 1| + V_2(\vec{r}_i, t)|2\rangle\langle 2| \) for the position and momentum \( \vec{p}_i, \vec{r}_i \) of the \( i \)th particle. The trapping field is arbitrary but the simplest case corresponds to harmonic traps: \( V_i = E_i + \frac{1}{2}m\omega_i r^2 \).

A base of the Hilbert space will be an ensemble of states \( \bigotimes_{i=1}^{N} |p_i, 1 \text{ or } 2⟩⟩ \otimes |\Pi_{k\sigma} n_{k\sigma}⟩⟩ \) when using the Fock notation for the field. We treat the \( N \) particles as totally independent and use the density matrix formalism (written as \( \hat{\rho} \)) to describe the system of \( N \) identical particles as a statistical ensemble. The external field is common to the \( N \) atoms and this can automatically generate entanglement between the atoms or collective behaviour that can indeed lead to cooling [55 56]. As explained in the article, this is not our interest here and we shall study only the single particle case. In the dipolar approximation and neglecting the Roentgen term, despite the fact that it can create surprising radiation forces on the atoms [50 57], the Hamiltonian for a single particle reads as:

\[
\hat{H} = \frac{\vec{p}^2}{2m} + V_1(\vec{r}, t)|1\rangle\langle 1| + V_2(\vec{r}, t)|2\rangle\langle 2| - d.\vec{E}(\vec{r}, t)(|2\rangle\langle 1| + |1\rangle\langle 2|) + \sum_{k\sigma} \hbar \omega_k \left( \hat{a}_{k\sigma} \hat{a}_{k\sigma}^{\dagger} + 1/2 \right) \tag{4}
\]

where \( d \) is the transition dipole element (assumed to be real \( d = (2|q|1) \)) and \( \vec{E}(\vec{r}, t) \) is a quantized real field. For instance for a single plane wave field (in a volume \( L^3 \)) \( \vec{E}(\vec{r}, t) = \sum_{k, \sigma} i \sqrt{\frac{\hbar \omega_k}{2\epsilon_0 L^3}} \left( \hat{a}_{k\sigma} e^{-i\omega_k t} \epsilon_{k\sigma}^* e^{ikr} - \hat{a}_{k\sigma}^{\dagger} e^{i\omega_k t} \epsilon_{k\sigma} e^{-ikr} \right) \).

The initial state is uncorrelated and density operator can be written as an atomic (external and internal degrees of freedom) and a field part as \( \hat{\rho} = \hat{\rho}_{\text{ext}} \otimes \hat{\rho}_{\text{field}} = \hat{\rho}_{\text{ext}} \otimes \hat{\rho}_{\text{int}} \otimes \hat{\rho}_{\text{field}} \).

In the semi-classical approximation, we would like to replace the field operators (denoted with the hat \( \hat{\cdot} \)) by their classical expectation values, namely \( \langle \hat{a}_{k\sigma} \rangle \) and \( \hat{a}_{k\sigma}^{\dagger} \) by c-numbers \( a_{k\sigma} \) and \( a_{k\sigma}^{\dagger} \), such as \( \vec{E}(\vec{r}, t) \) by \( \vec{E}(\vec{r}, t) \) becomes in the Hamiltonian

\[
\hat{H} = \frac{\vec{p}^2}{2m} + E_1(\vec{r}, t)|1\rangle\langle 1| + E_2(\vec{r}, t)|2\rangle\langle 2| - d.\vec{E}(\vec{r}, t)(|2\rangle\langle 1| + |1\rangle\langle 2|)
\]

Classical fields

This can be done, by using coherent states \(|\alpha⟩\rangle\), that are eigenstates of the annihilation operator \( \hat{a} \) : \( \hat{a}|\alpha⟩\rangle = \alpha|\alpha⟩\rangle \), by using the unitary transformation under the operator \( \hat{U} = \mathcal{D}(\alpha, e^{-i\omega \lambda}) \) and neglecting the quantum field that now describes spontaneous emission only [59 60].

Therefore, in the following we assume to have classical laser fields with different frequencies \( \omega_L \), wave-vectors \( k_L \) or temporal phase \( \Phi_L(t) \): \( \vec{E}(\vec{r}, t) = \vec{E}'(\vec{r}, t) + \vec{E}^f(\vec{r}, t) = \frac{1}{2} \sum_L \left[ E_L(t) e^{i(k_L \cdot \vec{r} - \omega_L t - \Phi_L(t))} + E_L^*(t) e^{-i(k_L \cdot \vec{r} - \omega_L t - \Phi_L(t))} \right] \).

The rotating wave approximation leads to

\[
\hat{H} = \frac{\vec{p}^2}{2m} + V_1(\vec{r}, t)|1\rangle\langle 1| + V_2(\vec{r}, t)|2\rangle\langle 2| - d.\vec{E}'(\vec{r}, t)|2\rangle\langle 1| - d.\vec{E}^f(\vec{r}, t)|1\rangle\langle 2| \tag{5}
\]
We will now use this Hamiltonian to describe the evolution. In matrix notation with the \([1,2]\) basis, the Hamiltonian \(\hat{H}\) becomes
\[
\hat{H} = \begin{pmatrix}
\hat{H}_1 & \hat{V} \\
\hat{V}^\dagger & \hat{H}_2
\end{pmatrix}
\]
where the coupling term is \(\hat{V} = -\frac{d}{2} \sum L E_L(t)e^{i(k_L r - \omega_L t - \Phi_L(t))} = \sum L \hat{V}_L\).

**Density matrix**

The time evolution \(i\hbar \frac{\partial \hat{\rho}}{\partial t} = \hat{H} \hat{\rho} - \hat{\rho} \hat{H}\) leads to:
\[
\begin{pmatrix}
\frac{\partial \rho_{11}}{\partial t} & \frac{\partial \rho_{12}}{\partial t} \\
\frac{\partial \rho_{21}}{\partial t} & \frac{\partial \rho_{22}}{\partial t}
\end{pmatrix} = \frac{1}{i\hbar} \begin{pmatrix}
[\hat{H}_1, \hat{\rho}_{11}] + \hat{V}^\dagger \hat{\rho}_{21} - \hat{\rho}_{12} \hat{V} + [\hat{p}^2/2m, \hat{\rho}_{12}] + \hat{V}_1 \hat{\rho}_{12} - \hat{\rho}_{12} \hat{V}_2 + \hat{V}^\dagger \hat{\rho}_{22} - \hat{\rho}_{11} \hat{V}^\dagger \\
[\hat{H}_2, \hat{\rho}_{21}] + \hat{V}_2 \hat{\rho}_{21} - \hat{\rho}_{21} \hat{V}_1 + \hat{V} \hat{\rho}_{11} - \hat{\rho}_{22} \hat{V}
\end{pmatrix}
\]

**Wigner functions**

The Wigner-Weyl transform of this equation gives the time evolution of the Wigner function defined as
\[
\rho_{12}(r,p,t) = \frac{1}{\hbar^3} \int (p' - p/2) \rho(\hat{r}, \hat{p}, t)|p + p'/2, e^{-ir\cdot p'/\hbar}, dp'
\]

through the so-called Moyal bracket, governed by
\[
\frac{\partial W}{\partial t} = \frac{1}{i\hbar} (H * W - W * H)
\]

The \(*\)-product can be evaluated using the convenient formula \([20]\) for any generic function \(\rho_{1,2}(r,p)\)
\[
(\rho_1 * \rho_2)(r,p) = \rho_1 (r, -\hat{p} - \frac{\hbar}{2} \frac{\partial}{\partial p}, \rho - i \frac{\hbar}{2} \frac{\partial}{\partial r}) \rho_2(r,p)
\]
\[
(\rho_2 * \rho_1)(r,p) = \rho_2(r, -\frac{\hbar}{2} \frac{\partial}{\partial p}, \rho + i \frac{\hbar}{2} \frac{\partial}{\partial r}) \rho_1(r,p)
\]

that we have restricted to a one dimensional motion for simplicity.

Therefore, when no \(\hat{r}, \hat{p} \) product are present in \(\hat{\rho} = \rho(\hat{r}, \hat{p})\), the Wigner(-Weyl) transform \(W_\rho(r,p,t)\) is the unmodified classical observable expression \(\rho(r,p)\). An important example is a conventional Hamiltonian, \(\hat{H} = \hat{p}^2/2m + \hat{V}(\hat{r}, \hat{t})\), for which the transition from classical mechanics is the straightforward quantization: \(W_H(r,p,t) = H(r,p,t) = \hat{p}^2/2m + \hat{V}(r,t)\).

The expressions containing \(e^{ikL \cdot \Phi}\) can be expanded by using exponential (Taylor) series that indicates \(e^{ikL(r \pm \Phi)} f(r,p,t) = e^{ikL \cdot r} f(r,p \mp \hbar kL/2,t)\). and finally using \(i\hbar \Omega_L(r,t) = dE_L e^{i(kL \cdot r - \omega_L t - \Phi_L(t))}\), we obtain:
\[
\begin{align*}
\left[\frac{\partial}{\partial t} + \frac{\partial}{\partial r} - \frac{1}{\hbar} \left[\hat{V}_1(r, i \frac{\hbar}{2} \hat{p}_r) - \hat{V}_1(r, -i \frac{\hbar}{2} \hat{p}_r)\right]\right] W_{11}(r,p,t) &= -\frac{1}{2i} \sum_L (\Omega_L^* (r,t) W_{21}(r,p + \frac{\hbar k_L}{2}, t) - \Omega_L(r,t) W_{12}(r,p + \frac{\hbar k_L}{2}, t)) \\
\left[\frac{\partial}{\partial t} + \frac{\partial}{\partial r} - \frac{1}{\hbar} \left[\hat{V}_2(r, i \frac{\hbar}{2} \hat{p}_r) - \hat{V}_2(r, -i \frac{\hbar}{2} \hat{p}_r)\right]\right] W_{12}(r,p,t) &= -\frac{1}{2i} \sum_L (\Omega_L^* (r,t) W_{22}(r,p + \frac{\hbar k_L}{2}, t) - \Omega_L(r,t) W_{11}(r,p + \frac{\hbar k_L}{2}, t)) \\
\left[\frac{\partial}{\partial t} + \frac{\partial}{\partial r} - \frac{1}{\hbar} \left[\hat{V}_2(r, i \frac{\hbar}{2} \hat{p}_r) - \hat{V}_2(r, -i \frac{\hbar}{2} \hat{p}_r)\right]\right] W_{21}(r,p,t) &= -\frac{1}{2i} \sum_L (\Omega_L(r,t) W_{12}(r,p + \frac{\hbar k_L}{2}, t) - \Omega_L^* (r,t) W_{21}(r,p + \frac{\hbar k_L}{2}, t)) \\
\left[\frac{\partial}{\partial t} + \frac{\partial}{\partial r} - \frac{1}{\hbar} \left[\hat{V}_1(r, i \frac{\hbar}{2} \hat{p}_r) - \hat{V}_1(r, -i \frac{\hbar}{2} \hat{p}_r)\right]\right] W_{22}(r,p,t) &= -\frac{1}{2i} \sum_L (\Omega_L(r,t) W_{21}(r,p + \frac{\hbar k_L}{2}, t) - \Omega_L^* (r,t) W_{22}(r,p + \frac{\hbar k_L}{2}, t))
\end{align*}
\]
\[
\begin{align*}
\frac{\partial W_{11}}{\partial t} \bigg|_{\text{spont}} &= \Gamma \int_{-p_r}^{p_r} \Theta(p') W_{22}(r, p + p') dp' \\
\frac{\partial W_{11}}{\partial t} \bigg|_{\text{spont}} &= -\frac{\Gamma}{2} W_{12}(r, p) \\
\frac{\partial W_{21}}{\partial t} \bigg|_{\text{spont}} &= -\frac{\Gamma}{2} W_{21}(r, p) \\
\frac{\partial W_{22}}{\partial t} \bigg|_{\text{spont}} &= -\Gamma W_{22}(r, p)
\end{align*}
\]

where \( \Theta(p') \) is the probability density distribution for the projection of spontaneous emission \( \Theta(p') = \frac{3}{8p_r} \left( 1 + \frac{p'^2}{p_r^2} \right) \) for a dipolar radiation pattern) on the atomic recoil momentum for \( p_r = \hbar k \).

Equation of motion of the Husimi distribution can be derived \([59–63]\) and present non-zero second term of the Liouville equation (similar to Eqs. (8)-(11)).

Connection with Liouville equation

In the absence of light fields, Taylor series expansion indicates that the evolution of the diagonal terms \( W_{ii} \) is given by:

\[
\frac{DW_{ii}}{Dt} = \frac{\partial W_{ii}}{\partial t} + \frac{p}{m} \cdot \frac{\partial W_{ii}}{\partial r} - \frac{\partial V_{i}}{\partial r} \cdot \frac{\partial W_{ii}}{\partial p} = \sum_{s \geq 1} \frac{\hbar^{2s}}{(2s + 1)!} \frac{\partial^{2s+1} V_{i}}{\partial r^{2s+1}} \frac{\partial^{2s+1} W_{ii}}{\partial p^{2s+1}}
\]

We recover the Liouville’s equation, \( \frac{DW_{ii}}{Dt} = 0 \), under the influence of the potential \( V \), but only for a quadratic potential \( V_i(r, t) = a(t) + b(t)r + c(t)r^2 \). However, when higher derivatives of \( V_i(r) \) are present, additional terms will give rise to diffusion and the quantum Wigner function gradually deviates from the corresponding classical phase space probability density. So a non-harmonic potential is a clear way to modify the Wigner phase space density. This argument also applies to the Husimi function.

Interaction picture: free evolution

The evolution of \( H_1(t) \) is given by the unitary time evolution operator \( \hat{U}_1(t) = e^{-i\int H_1(t)/\hbar} \). In matrix notation, the evolution operator is \( \hat{U}_0 = \begin{pmatrix} \hat{U}_1 & 0 \\ 0 & \hat{U}_2 \end{pmatrix} \). The interaction picture consists in defining a new density matrix \( \hat{\rho}^I(t) = \hat{U}_0^\dagger \hat{\rho}(t) \hat{U}_0(t) \), which evolves under the modified Hamiltonian \( \hat{H}^I = \hat{U}_0^\dagger \hat{H} \hat{U}_0 + i\hbar \frac{d\hat{U}_0^\dagger}{dt} \hat{U}_0 = \begin{pmatrix} 0 & \hat{V}^I \\ \hat{V}^I & 0 \end{pmatrix} \) where \( \hat{V}^I = \hat{U}_2 \hat{V} \hat{U}_1 \).

Because several laser frequencies are possibly present, the interaction picture is more appropriate than the Bloch rotating frame. The latter would imply to choose one laser frequency as a reference. The interaction picture removes this arbitrariness.

Density matrix

Using the momentum representation, where \( \hat{r} \) acts as \( i\hbar \partial_p \) on \( \psi(p) = \langle p|\psi \rangle \), we have \( e^{ikp|p\rangle = |p + \hbar k\rangle} \) We find

\[
\hat{V}^I|p\rangle = -\frac{1}{2} \sum_L |p + \hbar k_L\rangle \Omega_L e^{-i(\delta_L^p + t)}
\]

\[
\delta_L^{p\pm} = \omega_L - (E_2 - E_1)/\hbar - \frac{k_L}{m}(p \pm \hbar k_L/2)
\]
where $\hbar\Omega_L(t) = d_E L e^{-\Phi_L(t)}$ and $\delta p^\pm = \delta L^0 + \delta L^p(p) \pm \delta L^t$. The detuning $\delta L^0 = \omega_L - (E_2 - E_1)/\hbar$, the Doppler shift $\delta L^p(p) = -kL/p/m$ and recoil frequency $\delta L^t = -\hbar k L^2/2m$ appear naturally.

With $\dot{\rho}^I_{ij} = \hat{U}_i ^\dagger \dot{\rho}^I_{ij} \hat{U}_j$, the evolution reads as:

$$
\left( \frac{\partial\dot{\rho}^I_{11}}{\partial t} \frac{\partial\dot{\rho}^I_{21}}{\partial t} \frac{\partial\dot{\rho}^I_{12}}{\partial t} \frac{\partial\dot{\rho}^I_{22}}{\partial t} \right) = \frac{1}{4i} \sum L \left( \hat{V}^T \dot{\rho}^I_{21} - \dot{\rho}^I_{12} \hat{V}^T \dot{\rho}^I_{12} - \dot{\rho}^I_{11} \hat{V}^T + \dot{\rho}^I_{22} \hat{V}^T \right) \quad (14)
$$

Assuming there is no external field from now and using $\rho^{I,p}_{ij} = \langle p^I | \dot{\rho}^I_{ij} | p \rangle = e^{i(p^2-\tilde{p}^2)t/2m\hbar} e^{i(E_i-E_j)t/\hbar} \rho^{I,p}_{ij}$, the latter can be written as:

$$
\left( \frac{\partial\rho^{I,p}_{11}}{\partial t} \frac{\partial\rho^{I,p}_{21}}{\partial t} \frac{\partial\rho^{I,p}_{12}}{\partial t} \frac{\partial\rho^{I,p}_{22}}{\partial t} \right) = \frac{1}{2i} \sum L \left( \Omega^e L e^{i\delta_L^t + \delta L^p \rho_{12}^{I,p}(p+hk_L)p - \Omega^e L e^{i\delta_L^t - \delta L^p \rho_{12}^{I,p}(p-hk_L)p} - \Omega^e L e^{i\delta_L^t + \delta L^p \rho_{21}^{I,p}(p-hk_L)p - \Omega^e L e^{i\delta_L^t - \delta L^p \rho_{21}^{I,p}(p+hk_L)p} - \Omega^e L e^{i\delta_L^t + \delta L^p \rho_{22}^{I,p}(p-hk_L)p - \Omega^e L e^{i\delta_L^t - \delta L^p \rho_{22}^{I,p}(p+hk_L)p} - \Omega^e L e^{i\delta_L^t + \delta L^p \rho_{11}^{I,p}(p-hk_L)p - \Omega^e L e^{i\delta_L^t - \delta L^p \rho_{11}^{I,p}(p+hk_L)p} \right) \quad (15)
$$

**Wigner function**

It is quite convenient to use the so-called non-diagonal Wigner functions by defining $W^I_{ij} = W^I_{ij}/\hbar$ as the Wigner transform function associated to $\dot{\rho}^I_{ij} = \langle i | \dot{\rho}^I_{ij} | j \rangle$. So $W^I_{ij}(r,p,t) = e^{i(E_i-E_j)t/\hbar} W^I_{ij}(r-\tilde{p}t/m,p,t)$ and the evolution equations become:

$$
\frac{\partial W^I_{11}}{\partial t}(r,p,t) = -\sum L \left[ \Omega^e_L(r,p,t) W^I_{21}(r-hk_L t/2m,p+hk_L/2,t) \right] \quad (16)
$$

$$
\frac{\partial W^I_{21}}{\partial t}(r,p,t) = \frac{1}{2i} \sum L \left( \Omega^e_L(r,p,t)(W^I_{22}(r-hk_L t/2m,p+hk_L/2,t) - W^I_{11}(r+hk_L t/2m,p-hk_L/2,t)) \right) \quad (17)
$$

$$
\frac{\partial W^I_{22}}{\partial t}(r,p,t) = \sum L \left[ \Omega^e_L(r,p,t) W^I_{21}(r+hk_L t/2m,p-hk_L/2,t) \right] \quad (18)
$$

where

$$
\Omega^e_L(r,p,t) = \Omega^e_L e^{i(k_L r + \frac{hk_L t}{m} - \delta_L^t - \Phi_L(t))} \quad (19)
$$

**Single laser case (Bloch equation)**

When there is only one laser, we can define

$$
\hat{W}^I_{ij}(r,p,t) = W^I_{ij}(r,p,t)
$$

$$
\hat{W}^I_{11}(r,p,t) = W^I_{11}(r,p,t)
$$

$$
\hat{W}^I_{22}(r,p,t) = W^I_{22}(r-hk_L t/m,p+hk_L,t)
$$

$$
\hat{W}^I_{21}(r,p,t) = e^{-i(k_L r + \frac{hk_L t}{m} - \delta_L^t - \Phi_L(t))} W^I_{21}(r-hk_L t/2m,p+hk_L/2,t) \quad (20)
$$

If we assume $\Omega^e_L$ real, the evolution is governed by

$$
\frac{\partial}{\partial t} \hat{W}^I_{11} - \hat{W}^I_{22} = \left( -\Omega_L + \frac{hk_L}{2m} \frac{\partial}{\partial r} \hat{W}^I_{22} \right) \quad (21)
$$

$$
\frac{\partial}{\partial t} \hat{W}^I_{22} - \hat{W}^I_{11} = \left( \Omega_L - \frac{hk_L}{2m} \frac{\partial}{\partial r} \hat{W}^I_{21} \right) \quad (22)
$$

$$
\frac{\partial}{\partial t} \hat{W}^I_{21} = \hat{W}^I_{22} \quad (23)
$$
We recognize the standard Bloch equations except for the term in \( \frac{\hbar k}{2m} \frac{\partial}{\partial r} \). We can thus retrieve the Bloch equations from the exact Wigner function evolution by performing series expansion in \( \hbar k \). This approach justifies the semi-classical equation for the particles evolution that we derive from heuristic considerations.

**SEMI-CLASSICAL EVOLUTION**

From the quantum evolution, we can derive the semi-classical evolution of the atomic motion. The underlying assumption is that the displacement of the atom during the internal relaxation time is very small. The internal variables follow quasi-adiabatically the external motion [19]. It is then possible to separate the internal and the external degree of freedom.

The Doppler or recoil effects, or the use of the stationary state of the Bloch equation can be done with hand-waving arguments (see for instance in Ref. [64]). Nevertheless, the Lagrangian description (individual particles are followed through time), Eulerian description and interaction picture that freeze the motion in the Eulerian description may lead to confusion. We will clarify this distinction.

**Definition of a force**

For simplicity, we neglect the external potentials (but they can be included in the interaction picture if needed).

In the semi-classical approach, the particle motion is classical: for a given particle initially at \( r(t_0) = r_0 \) and \( v(t_0) = v_0 \) at time \( t = t_0 \) its trajectory in phase space \( r(t), v(t) \) is given by Newton’s equation of motion \( m \frac{dv}{dt} = F(r(t), v(t), t) \).

The standard way to define the force in laser cooling is by using the Ehrenfest theorem (see for instance [65, 66], but other methods exists [67,69]). Knowing the light field seen by the atom at the position \( r \) with velocity \( v = p/m \) enables to solve the optical Bloch equations (density matrix \( \hat{\sigma}(t) \) evolution) to determine the atomic internal state. The force is then derived from \( F = -tr[\hat{\sigma}(t) \nabla \hat{H}] \). The usual optical Bloch equations where \( \sigma_{ij}(t) \) stands for \( \sigma_{ij}(t; r_0, v_0, t_0) \) read as

\[
\left( \frac{\partial \sigma_{11}}{\partial t}, \frac{\partial \sigma_{12}}{\partial t}, \frac{\partial \sigma_{21}}{\partial t}, \frac{\partial \sigma_{22}}{\partial t} \right) (t) = -\frac{1}{2i} \sum_{L} \begin{pmatrix}
\Omega_1^+(r(t), t) \sigma_{21}(t) - \Omega_1^-(r(t), t) \sigma_{12}(t) & \Omega_2^+(r(t), t) (\sigma_{12}(t) - \sigma_{21}(t)) \\
\Omega_1^-(r(t), t) (\sigma_{11}(t) - \sigma_{22}(t)) & \Omega_2^+(r(t), t) \sigma_{12}(t) - \Omega_2^-(r(t), t) \sigma_{21}(t)
\end{pmatrix}
\]

where \( \Omega_1^+(r, t) = \Omega_1 e^{i(\omega t - r \cdot \omega L t - \Phi_L(t))} \). The rapidly oscillating terms can be removed by introducing slowly varying quantities as \( \sigma_{ij}^\prime(t) = e^{-i(E_i - E_j) t / \hbar} \sigma_{ij}(t) \).

The absence of Doppler shift in the expression of \( \Omega_L(r, t) \) may be surprising, especially when compared to Eq. (15) (using \( \beta = p = p(t), r = r(t) \) and \( \hbar k_L \) put to 0). The explanation is the following: we use \( r(t) = r(t; r_0, v_0, t_0) \) so the Lagrangian description where individual particles are followed through time, whereas, when dealing with the Wigner \( W(\mathbf{r}, \mathbf{v}, t) \) or PSD \( \rho(\mathbf{r}, \mathbf{v}, t) \) picture, we use in the Eulerian description. The connection between Lagrangian and Eulerian coordinates explains why the Doppler effect is correctly taken in both Eq. (23) with \( \Omega_1^+(r(t), t) = \Omega_L e^{i(k_L r(t) - \omega(t) t - \Phi_L(t))} \), and in Eq. (15) with \( \Omega_L(r, p, t) = \Omega_L e^{i(k_L r + k_L p t/m - \Phi_L(t))}. \) In any case, the instantaneous laser phase seen by the atoms is correct, including the Doppler effect because \( \frac{dr(t)}{dt} = p(t)/m \).

Similarly, in the Eulerian description the force is thus given by \( \text{Tr}[\hat{\sigma}(t) \nabla \hat{H}] \), or \( \text{Tr}[\hat{p}(t)^I \nabla \hat{V}^I] \) using the cyclic invariant of the trace. We have \( V^I(r, p, t) = -\sum_L \hbar \Omega_L(r, p, t) \) so \( \nabla V^I(r, p, t) = -i \sum_L \frac{\hbar k_L}{2} \Omega_L(r, p, t) \). So in conclusion and back to our Lagrangian description we have:

\[
F(r(t), v(t), t) = \Im \left[ \sigma_{21}(t) \sum_L \hbar k_L \Omega_L^+(r(t), t) \right]
\]

As we chose plane waves (or \( \nabla E_L = 0 \)), there is no direct dipolar force. Also, because of the interplay between the Bloch equations (Eq. [23]) and the force (Eq. [24]), the atomic velocity \( v(t) \) and position \( r(t) \) should be updated in a short time interval (typically ps), and the calculation of the Bloch equation evolution iterated on a similar time scale [64].
Phase space evolution equation

Here, we would like to justify the equations we just derived assuming a separation of the external and internal degrees of freedom. However, we know that without spontaneous emission, this is valid only if the ratio of resonant photon momentum to atomic momentum dispersion is small $\hbar k/\Delta p \ll 1$. In such a case, the rapid processes acting on the internal degrees of freedom can be separated from the slow processes associated with translational motion. The dynamics of the atomic ensemble is thus determined by the slow change of the distribution function in translational degrees of freedom $w(r, p) = W_{11} + W_{22}$ and the expansion in $\hbar k$, that we will derive here for completeness, is justified.

One analogue of the classical phase space distribution is the total distribution function in translational degrees of freedom, $w(r, p, t)$ as plotted in [55 Fig. 2(b)]. Equations [8,11] (written for simplicity without the external potentials), become:

$$\frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} W_{11}(r, p, t) = - \frac{1}{2i} \sum_L \left[ \Omega_L^*(r, t) W_{21}(r, p + \hbar k_L/2, t) - \Omega_L(r, t) W_{12}(r, p + \hbar k_L/2, t) \right]$$  \hspace{1cm} (25)

$$\frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} W_{12}(r, p, t) = - \frac{1}{2i} \sum_L \left[ \Omega_L^*(r, t) W_{22}(r, p + \hbar k_L/2, t) - W_{11}(r, p - \hbar k_L/2, t) \right]$$  \hspace{1cm} (26)

$$\frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} W_{21}(r, p, t) = - \frac{1}{2i} \sum_L \left[ \Omega_L(r, t) W_{11}(r, p - \hbar k_L/2, t) - W_{22}(r, p + \hbar k_L/2, t) \right]$$  \hspace{1cm} (27)

$$\frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} W_{22}(r, p, t) = - \frac{1}{2i} \sum_L \left[ \Omega_L(r, t) W_{12}(r, p - \hbar k_L/2, t) - \Omega_L^*(r, t) W_{21}(r, p - \hbar k_L/2, t) \right]$$  \hspace{1cm} (28)

with $\hbar \Omega_L(r, t) = dE_p e^{i(k_{\perp} \cdot r - \omega_{\perp}(t - \Phi_0))}$.

An frequently used method to derive a continuity equation as [55 Eq.(1)] for $\rho = w$ is to expand the Wigner distribution equations in a power series of the photon momentum $\hbar k$. [19,58,70–74]. In the presence of spontaneous emission, the second order leads to the standard Fokker-Planck equation [19,58,70–74]. The simplest formulation is restricted to the first order approximation, therefore $W_{21}(r, p + \hbar k_L/2, t) \approx W_{21}(r, p, t) + \frac{\hbar k_L}{2} \frac{\partial}{\partial p} W_{21}(r, p, t)$. To this first order in $\hbar k_L$, the sum of [25] and [28] is:

$$\left[ \frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} \right] w(r, p, t) = - \sum_L \Im \left[ \Omega_L^*(r, t) \hbar k_L \frac{\partial}{\partial p} W_{21}(r, p, t) \right]$$  \hspace{1cm} (29)

Since the recoil momentum $\hbar k$ is small, the variation of atomic translational motion is slower than the atomic internal state change. The latter follows the varying translational state $w(r, p, t)$ [75]. Fast relaxation of the internal atomic state means that, the functions $W_{ij}(r, p, t)$ follow the distribution function $w(r, p, t)$.

At zero order in $\hbar k_L$ we have the simplest approximation $W_{ij}(r, p, t) \approx W_{ij}^0(r, p, t) w(r, p, t)$. Eq. [29] leads to

$$\left[ \frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} \right] w(r, p, t) = - \frac{\partial}{\partial p} [F(r, p, t) w(r, p, t)]$$  \hspace{1cm} (30)

We recognize a continuity equation as [55 Eq.(1)] with the force given by

$$F(r, p, t) = \Im \left[ W_{21}^0(r, p, t) \sum_L \hbar k_L \Omega_L^*(r, t) \right]$$  \hspace{1cm} (31)

So in a classical picture, this expression of the force shall be used to calculate individual particles trajectories. The evolution of the Wigner function is given by Eqs. [25]:[28], with $W_{ij}(r, p, t) \approx W_{ij}^0(r, p, t) w(r, p, t)$, to obtain

$$\frac{\partial W_{11}^0(r + pt/m, p, t)}{\partial t} = - \sum_L \Im \left[ \Omega_L^*(r + pt/m, t) W_{21}^0(r + pt/m, p, t) \right]$$  \hspace{1cm} (32)

$$\frac{\partial W_{21}^0(r + pt/m, p, t)}{\partial t} = \frac{1}{2i} \sum_L \Omega_L(r + pt/m, t) (W_{22}^0(r + pt/m, p, t) - W_{11}^0(r + pt/m, p, t))$$  \hspace{1cm} (33)

$$\frac{\partial W_{22}^0(r + pt/m, p, t)}{\partial t} = \sum_L \Im \left[ \Omega_L^*(r + pt/m, t) W_{21}^0(r + pt/m, p, t) \right]$$  \hspace{1cm} (34)
where we have used \[ \frac{\partial}{\partial t} + \frac{p}{m} \frac{\partial}{\partial r} \] \[ W_{11}^0(r + pt/m, p, t) = \partial W_{11}^0(r + pt/m, p, t) \].

We partially recognize the optical Bloch equations (Eqs. 23), with \( \sigma_{ij}(t) = W_{ij}^0(r_0 + p_0t/m, p_0, t) \) \[19\]. This is the usual first order in time connection between Lagrangian and Eulerian specification: \( r(t) = r(t; r_0, v_0, t_0) \approx r_0 + v_0t, p(t) \approx p_0 \). So to first order \( \sigma_{ij}(t) \approx W_{ij}^0(r(t), p(t), t) \) and the force given by Eq. (31) is exactly the same force as Eq. (24).

An alternative way to derive these expressions consists in using the interaction picture. A similar method using \( w^I(r, p, t) = W_{11}^I + W_{22}^I W_{11}^I W_{11}^I \approx W_{11}^I(r, p, t)w(r, p, t) \) from Eqs. (16)–(18) leads to first order in \( \hbar k_L \) to:

\[
\frac{\partial W_{11}^I}{\partial t}(r, p, t) = - \sum L \Im \left[ \Omega^*_L(r, p, t) W_{21}^I (r, p, t) \right] \\
\frac{\partial W_{21}^I}{\partial t}(r, p, t) = \frac{1}{2i} \sum L \Omega_L(r, p, t) \left( W_{22}^I (r, p, t) - W_{11}^I (r, p, t) \right) \\
\frac{\partial W_{22}^I}{\partial t}(r, p, t) = \sum L \Im \left[ \Omega^*_L(r, p, t) W_{21}^I (r, p, t) \right]
\]

which are the usual Bloch equations in the particle frame. The Doppler effect is here explicitly included. Indeed, the continuity equation reads as

\[
\frac{\partial w^I}{\partial t}(r, p, t) = - \left[ \frac{t}{m} \frac{\partial}{\partial r} + \frac{\partial}{\partial p} \right] \left( F^I(r, p, t) w^I(r, p, t) \right)
\]

for the force \( F(r + pt/m, p, t) = F^I(r, p, t) = \sum L \hbar k_L \Omega^*_L(r, p, t) W_{21}^I (r, p, t) \).

This is indeed the classical continuity equation \[55\] Eq. (1)]. In the interaction picture \( \rho(r, p, t) = \rho^I(r - pt/m, p, t) \) leads to

\[
\frac{\partial \rho^I}{\partial t}(r, p, t) + \left[ \frac{t}{m} \frac{\partial}{\partial r} + \frac{\partial}{\partial p} \right] \left( \rho^I F^I(r, p, t) \right) = 0
\]

where \( F(r, p, t) = F^I(r - pt/m, p, t) \).
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