Optical gratings in the collective interaction between radiation and atoms, including recoil and collisions

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(Received 15 March 2001, revision received 10 June 2001)

Abstract. The introduction of collisions and of a thermal distribution for the atomic momentum in the model for the Collective Atomic Recoil Laser (CARL) is at the origin of important modifications in the interpretation of the mechanisms that give rise to the amplification of the backreflected wave. It is shown that the atomic density grating, considered to be the cause of gain in CARL, disappears in the presence of collisions, while other gratings—in population and polarization phase—survive. While the population grating appears to be merely a consequence of the collective interaction, the latter is the likely cause for the instability. Finally, simulations show that models that make use of an exponential relaxation mechanism for the atomic momentum, rather than accounting for collisions explicitly, largely overestimate the strength of the interaction.

1. Introduction

The quasi-resonant interaction between atoms and strong electromagnetic fields has been the subject of intense investigations for the last forty years. In the course of these studies, a wide range of effects has been identified and the various causes have enlivened the field of nonlinear atomic spectroscopy for many years.

In most investigations, except for very early ones [1], the atomic recoil in light-matter interaction has been neglected. Two reasons justify this assumption: the atomic momentum conservation is automatically satisfied, and—for thermal atoms at room temperature—the change in momentum due to the single interaction is negligible compared with the average atomic momentum (typically some four orders of magnitude lower than that of the most populated velocity classes). Systems where the atomic sample behaves as an ensemble, such as lasers, have been traditionally modelled without taking into account the individual atomic recoil effect. The rationale behind this assumption is that the comparison between experimental observations and models which did not take into account atomic recoil provided very satisfactory results. The likely reason for this is that the intrinsic disorder in the external atomic degrees of freedom, proper for a thermal sample, masks the possible influence that recoil may have on the global behaviour.
once ensemble averages are performed. Indeed, the instability that gives rise to the coherent emission takes place in the internal degrees of freedom, with little influence coming from the external ones (summarized in a certain amount of Doppler broadening). In the realm of atomic spectroscopy, however, recoil has been playing an ever increasing role, since cooling techniques have become more and more widespread. Mechanical effects of light on atoms were investigated early on [2–4] and their influence on atomic resonances, the so-called recoil-induced resonances, has been widely studied both theoretically [5] and experimentally [6].

For systems displaying a collective interaction with light, the question of the introduction of recoil has naturally arisen in a Free Electron Laser [7]. There, the electrons are (nearly) monokinetic and it is therefore natural to expect that, in the reference system of the centre of mass, the momentum transfer from the field–electron interaction may give average results that add up with a certain degree of coherence, thereby giving rise to observable effects. By extension, the possibility was suggested that recoil effects could appear also in atomic systems [8] where a collective behaviour may lead to the spontaneous formation of a density grating, a sort of ‘moving Bragg mirror’ (the so-called CARL system). Following this first prediction, several theoretical papers have dealt with different aspects of recoil in the nonlinear collective response between atoms and fields, in particular as far as optical bistability is concerned [9], and active or passive optical cavities [10]. Finally, a connection between the single-atom recoil and the collective behaviour has been theoretically investigated in [11].

The experimental verification of the influence of recoil in the collective behaviour of atomic samples has shown effects which were not traditionally attributed to the recoilless interaction [12, 13]. This seems to underline the importance of recoil in a collective interaction. However, direct observations of a density grating (considered to be the signature of the collective interaction) are very difficult, since several other kinds of grating can coexist and superpose, thereby making the identification of the signature of a pure density grating quite difficult. Because of this fact, the interpretation of the experimental results is not univocal and a theoretical alternative explanation, based on the presence of an atomic polarization grating, was proposed [14]. The validity of such alternative mechanism has been confirmed in an ad hoc experiment, designed at highlighting the influence of polarization gratings in a pump-probe experiment [15]. In spite of the validity of this interpretation key [14], measurements taken in the same setup as in [12], but in the weak probe limit [16] (i.e. where recoil effects could be neglected in the collective hot system, while the polarization grating could still play a role), have shown that the phenomenology is different from that observed for strong pumping [12]. It is therefore still legitimate to suspect that recoil may play a nonnegligible role in the observations of [12, 13].

The main difference between the experimental situation and the models used so far to describe the collective interaction in the presence of recoil [8–10], lies in the fact that a (nearly) monokinetic beam of (neutral) atoms cannot be obtained with a high enough density and that collisions with a buffer gas, necessary for technical reasons in most setups, are not taken into account. These two points represent a strong obstacle to an interpretation of the experimental results in terms of the collective interaction between e.m. field and atoms proposed in [8]. Indeed, the predicted existence of a density grating becomes immediately doubtful if one considers first the perturbing effect of collisions between optically active atoms and
buffer gas, and secondly the large width of the atomic momentum distribution.† This latter point is all the more important owing to the fact that experiments have been conducted so far in hot samples (≈500K or beyond—the only experiment using cold atoms has been performed in a Bose–Einstein condensate [17], where the physical interpretation is much simpler, since the matter is already in a coherent state).

In this paper, we discuss some further points arising from first-principles modelling [18], where we add to the original model [8] collisions with thermally distributed buffer gas atoms. In particular, we concentrate on three different aspects: the analysis of the different kinds of gratings that appear in the system, the reorganization of the atomic velocity distribution in the presence of the collective interaction, and the degree of coherence that the two main different modelling choices, [18] and [8], introduce, thereby strongly influencing the outcome of qualitative and quantitative predictions.

The original model [8] (formally introduced here below), includes in a standard way the phenomenological relaxation mechanisms for the internal degrees of freedom of the atoms. However, no relaxation is introduced for the translational degrees of freedom. While this approximation was not crucial in the first numerical investigations, since they were directed at studying only the transient behaviour, for the long-term dynamics a paradox would occur: the atoms would be accelerated without any bounds. As a refinement of the original model [8], an exponential relaxation (as well as an initial Gaussian distribution) of the atomic velocities towards a steady state was assumed in a connected paper [19], which treated all atoms in the same way (see [20] for a detailed physical discussion on this point). As we will see, the assumption of an exponential relaxation artificially introduces a much stronger degree of coherence among atoms, thereby enhancing the strength of the asymptotic collective behaviour, while lengthening the transient buildup.

The microscopic model introduced in [8] involves four variables to describe each atom: the complex polarization $S_j$, the population inversion $D_j$, the position $\theta_j$ and the momentum $P_j$. Additionally, there is a complex equation for the output (back-propagating with respect to the pump) field: $A_1$. The dynamics of the input (pump) field $A_2$ is neglected, as the model equations are derived under the approximation of a weak response. Figure 1 presents the setup described by this model. The equations from [8] read as:

$$
\dot{\theta}_j = P_j, \\
\dot{P}_j = 2\text{Re}[-A^*_1e^{-i\theta_j} + A^*_2S_j], \\
\dot{S}_j = \frac{i}{2}(P_j + 2\Delta_{20})S_j - \rho D_j(A_1e^{i\theta_j} + A_2) - \Gamma S_j, \\
\dot{D}_j = 4\rho \text{Re}[(A^*_1e^{-i\theta_j} + A^*_2)S_j] - \Gamma (D_j - D_{eq}), \\
\dot{A}_1 = i\Delta_{21}A_1 + \frac{1}{N}\sum_{j=1}^{N}S_je^{-i\theta_j}. 
$$

†Despite the change in the shape of the initial thermal momentum distribution, due to the light-matter interaction, it was observed that its width remains of same order of magnitude.
The slow variables $A_1$ and $S_j$ have been introduced so as to take $\omega_2$ as a reference for the field frequencies. The corresponding pump amplitude $A_2$ is a constant (parameter) of the model. Time has been scaled by $\tau = \omega_r \rho$ ($\omega_r$ being the single-photon recoil frequency shift), momentum by $\hbar k \rho$, and position by $(2k)^{-1}$. $\Gamma$ is the normalized atomic decay rate, which has been assumed identical for diagonal and off-diagonal density matrix elements. $\rho$ is the CARL parameter, related to the atomic density in the cell. $\Delta_{20}$ and $\Delta_{21}$ are the detunings of the input field frequency relative to the atomic and the output field frequencies, respectively. Here, the interaction of a passive medium with radiation is modelled. Consequently, the equilibrium population inversion $D_{eq}$ is fixed at the ground state.

In order to describe the collisions accurately, the interaction between optically active atoms and a heat bath is included in a microscopic way. Besides the deterministic evolution described by the set of equations (1), it is assumed that each atom independently undergoes random collisions whose effect is to reset its momentum to a Gaussian distributed value and its polarization phase to a uniformly distributed value in the interval $[0, 2\pi]$. It is assumed that the buffer gas does not interact with the light, and that its density is much higher than the density of optically active atoms. Given these two realistic hypotheses, it is considered that the momentum distribution of the buffer gas is not modified. Moreover, the rare collisions which involve two optically active atoms, or more than two atoms, are neglected.

The mean free path of a buffer gas atom in the optically active medium is $l = 4/[\pi (d_1 + d_2)^2 n]$ [21], where $d_1$ and $d_2$ are the diameters of buffer gas and optically active atoms, respectively, and $n$ is the density of the optically active medium. For the parameter values used in the simulations, and also under the experimental conditions [12, 13, 15], $l$ is not smaller than the size of the cell (of the order of $10^{-2}$ m). For example, assuming $d_1 = d_2 = 10^{-10}$ m, one obtains $l = 3$ m for $n = 9.8 \times 10^{18}$ m$^{-3}$, the density used in the experiment of [12]. In the present simulations, the density $n$ is smaller and, thereby, $l$ is larger. Hence, momentum and position distributions for the buffer gas atoms are mainly determined by collisions with the cell walls.

In addition, the experiments were conducted under conditions for which the number of buffer gas atoms is $10^2$ to $10^4$ times larger than that of optically active ones. Therefore, one does not expect substantial modifications of the equilibrium distribution of the buffer gas atoms owing to collisions with the active ones (which are out of thermodynamical equilibrium).

It is only for very high values of the atomic density ($n_{lim} = 8.9 \times 10^{20}$ m$^{-3}$, corresponding to normalized density values $\rho = 10^4$) that modifications of the
buffer gas velocity distribution should be taken into account. However, this regime is far beyond that considered in this paper.

Parameter values for \( \rho \), \( A_2 \), and detunings were set to the same orders of magnitude as those previously chosen \([8, 18, 22]\). More precisely, the simulations discussed here were performed with \( \Gamma = \Delta_{21} = 1 \), \( \Delta_{20} = -15 \) and \( \rho = 10 \). Thus, the normalized polarization damping constant \( \Gamma \) corresponds to a value of 6.3 \( \mu s \)\(^{-1} \) in physical units. For the (new) collision parameters, an exponential distribution of the time between two consecutive collisions, \( t_c \), has been assumed for each atom. The average of \( t_c \) for a given optically active atom (which is related to the total pressure) has been chosen to be \( T_c = 48 \) (corresponding to 7.2 \( \mu s \) in physical units), while the variance of the momentum distribution has been fixed at \( \sigma^2 = 33.3 \), which, with the above normalizations, corresponds to a temperature of 7 mK. Although these results were obtained by considering a cold buffer gas, contrary to the common experimental conditions \([12, 13, 15]\), similar behaviour can be observed at much higher temperatures.

The dynamics are modelled as self-generated output oscillations, i.e. the probe field starts from a tiny initial value (for example \( A_1^{\text{ini}} = 10^{-10} \)), so as to simulate the effect of an initial fluctuation corresponding to spontaneous emission. Initially, the atoms are randomly distributed in space with a Gaussian momentum distribution. From these initial conditions the system evolves towards a steady state, where all the ensemble averages, the output field amplitude, and the optical frequency are constant. It was observed that a steady condition would be attained for all parameter sets (\( \rho, A_2 \), detunings) used.

Depending on the pump intensity, two different regimes of operation were found. In the former one, the output intensity decreases to 0 with increasing the number \( N \) of atoms. This testifies to an incoherent emission of the atoms. In the latter regime, the average output intensity is independent of \( N \) (if \( N \) is large enough). For the choice of parameter values mentioned above, it was found that the critical (threshold) intensity where the phase transition occurs is \( I_c \approx 1.17 \pm 0.02 \) \([18]\).

In the original model \([8]\), the density grating has been identified as the main cause for the generation of a backpropagating electromagnetic field. This observation was quantified by the so-called bunching parameter, \( b \), which represents the spatial distribution of the atoms within a wavelength of the sinusoidal potential created by the superposition of the pump and the backpropagating fields \([8]\):

\[
b = \frac{1}{N} \sum_{j=1}^{N} e^{-i\theta_j}.
\]

In \([8]\) it was observed that this quantity is strongly different from zero. Under appropriate conditions, its maximum even approaches the theoretical limit (\( b = 1 \)). On the contrary, \( b \) is almost always negligible in all of our simulations. More precisely, it was observed that \( b \) converges exponentially to zero with a characteristic time of the order of \( T_c \). Hence, it is seen that random collisions are able to wash out any spatial structure. This fact is not a surprise in itself, as both collisions and the thermal velocity distribution of the atoms are expected to remove any inhomogeneity in the spatial distribution of atoms; such considerations have been at the origin of our attempt at modifying the model. However, one may be surprised to observe that at a temperature as low as 7 mK, there is still no
bunching. It is reasonable to expect that there would be a limit temperature (for a given pressure of buffer gas) above which \( b = 0 \) (despite the existence of an amplification\(^\dagger\)). Indeed, two competing processes take place simultaneously: coherent light–matter interactions which order the atoms in space, and random collisions which destroy this order. The limit mentioned above corresponds to the point where both these ordering and disordering effects compensate. It seems reasonable to imagine that below this temperature and pressure limit, one could observe a nonzero bunching. However, this situation appears to be extremely far from the usual experimental conditions (if one excludes a Bose–Einstein condensate, where the atomic sample has to be treated quantum-mechanically) and confirms the validity of the fundamental objections which can be raised to an interpretation of the experimental results [12, 13] solely in terms of [8].

In spite of the lack of a density grating, and hence of the apparent mechanism that is considered responsible for the CARL amplification, numerical simulations show gain in the direction opposite to the pump [18]. Hence, one should look for possible alternative sources of gain. It is easy to suspect that a likely cause should hide in the driving term of the output field evolution, and therefore the choice of a new parameter imposes itself in the form:

\[
c = \frac{1}{N} \sum_{j=1}^{N} S_j e^{-i\theta_j}.
\]

This coherence parameter \( c \) represents the correlation between internal and external degrees of freedom (\( S_j \) and \( e^{-i\theta_j} \)). While the spatial bunching \( b \) remains close to zero, \( c \) can attain a finite value, provided that the single-atom polarization and spatial phase synchronize. It is therefore this quantity that will reveal whether another source term may appear in the collective system.

As already mentioned, numerical simulations which include collisions and atomic momentum spread do not show any atomic bunching. In addition, they do not even show any evidence of a strong polarization grating\(^\ddagger\), and hence even the second proposed phenomenon [14] is unlikely to be the actual source of probe amplification. Instead, the presence of two other strong gratings was observed: one in the population, figure 2, and one in the phase of the atomic polarization, figure 3 (consider any one of the curves displayed, but one only at this point).

A careful look at figure 2 shows the presence of two main modulated structures. When the interaction strength in the vapour is large enough to observe a collective phenomenon, two counterpropagating e.m. waves are set up in the cell. The spatial modulation of the e.m. field resulting from their interference must therefore induce a variation in the level occupation of the atoms, depending on their position. Hence, an inversion grating should accompany any collective effect, and the appearence of the strong modulation (figure 2) is not surprising at all.

\(^\dagger\)The same qualitative behaviour (existence of a threshold for the pump intensity) was observed with \( T = 400 \text{K} \), and the same buffer gas pressure (corresponding to \( t_c = 48 \)). Above threshold, it was also noticed that \( b = 0 \).

\(^\ddagger\)By ‘polarization grating’ here is meant a spatial modulation of the total polarization (including its amplitude). A weak grating in the atomic polarization amplitude can be observed above threshold, but its structure is not very regular. Once the counterpropagating wave is amplified, an interference pattern is set up between the two fields and therefore all the internal variables must display, at least to a certain degree, a periodic spatial modulation.
Above threshold, the atoms gather mainly in two velocity groups (figure 4) (consider a curve for any value of $\Delta_{21}$, but only one at this point). It was observed that each one of the two structures in figure 2 corresponds to one of these two velocity groups.

The second strong grating that we find concerns the phase of the polarization (figure 3, one curve). Considering the structure of the model, its existence is not all
that surprising and sheds some light on another important physical mechanism that is active in the collective system. The driving term of $A_1$ is not the modulus of the atomic polarization, but the average of the (complex) polarization variable multiplied by a position factor. Hence, it becomes clear how the driving term in this system could be generated by a locking between the phase of the atomic polarization and the atom's position.

One might be surprised by the existence of a grating in the phase of the atomic polarization when phase-destroying collisions are explicitly taken into account in the model. A likely cause for the survival of such a grating is that the relaxation time of the polarization is smaller than the mean time between two collisions ($\tau_c \approx 50 \times \Gamma$). Hence, it is plausible to expect that the grating would have the time to reconstruct itself in spite of the perturbation. One would certainly expect the phase grating to disappear if the collisions become frequent enough. What is more interesting is the fact that the grating in the polarization amplitude is weak (and noisy), while collisions do not affect directly the polarization modulus. Although we do not have a definite answer to this point, there seems to be some strong evidence for phase locking that is insensitive to the strength of the modulus of the vector (polarization).

One more interesting piece of information concerning the gratings, which is clearly visible looking at the phase grating, is contained in the full figure 3. The different symbols show the datasets representing the phase of the polarization at different times. It can be seen that such a grating moves as a whole, following the sliding interference pattern of the two counterpropagating waves (due to their mutual detuning $\Delta_21$). Repeating the simulation, it was noticed that the grating velocity varies linearly with $\Delta_21$, from $\Delta_21 = -5$ to $\Delta_21 = 3$, thereby confirming the above remark. The shape of the structure does not change, which implies that the atoms can instantaneously adapt the phase of their polarization to their position.
in space. Thus, it is reasonable to expect that they are also capable of following the moving interference pattern.

This brings us to examination of the next point: the distribution of the atomic momenta in the coherent regime where the system acts as a whole. By looking at the dynamics of the average atomic momentum, one simply sees the mean effect of the radiation pressure on the atoms. The steady-state value is negative, which highlights the presence of a global recoil. We can learn a lot more by studying the distribution around this mean value.

Above the threshold value for the pump field, the shape of the distribution is clearly non-Gaussian (cf. figure 4, any curve). We now propose a mechanism which explains why. The total field amplitude in the cell and the phase of the atomic polarization depend on position (figure 3). This is also the case of the atom–field interaction potential \( V(\theta_j) = -E_j S_j \). Consequently, atoms interact with moving potential wells. If the pump intensity is set so as to obtain a steady state far above the threshold, the corresponding potential wells will be deep enough to trap some atoms, which will remain trapped until a strong enough collision ejects them. During this time, they are forced to move at the velocity of the grating. Thus, there is a privileged momentum class which should contain many more atoms than the others. This explains the peak in the momentum distribution.

For a given value of \( N \), the area under the distribution is fixed and should remain the same when one changes any parameter. It is therefore logical to notice that the peak which appears above threshold is accompanied by a depleted zone. Trapped atoms find themselves in a rather stable condition and therefore have a small probability of interacting with the field. Hence, they will not recoil as effectively as atoms that are outside the wells. On the contrary, atoms that are not moving at the same speed as the grating are liable to exchange momentum with the field, and this is particularly true for those atoms that are moving a bit slower than the grating. These velocity classes tend to lose more atoms, which recoil towards lower \( p \)-values than they can receive from the higher-lying classes, which coincide with the privileged velocity (peak of the distribution). Consequently, these classes are less populated and a hole appears on the left-hand side of the peak.

The plausibility of this mechanism was checked by plotting the momentum distribution for several values of \( \Delta_{21} \). Since it is known that the grating velocity varies linearly with \( \Delta_{21} \), both the peak and the hole can be expected to be shifted linearly as a function of \( \Delta_{21} \). This is exactly what appears in figure 4. Going from \( \Delta_{21} = 1 \) to \( \Delta_{21} = 5 \), one sees that the height of the peak decreases, while its width increases. This observation can be interpreted on the basis of the fact that the output intensity on steady state decreases, while the depth of the phase grating does not vary significantly. Hence, the trapping potential becomes shallower.

This description applies rather well to the simulations made for positive values of \( \Delta_{21} \). The shape of the momentum distribution becomes more complex for negative values of \( \Delta_{21} \). The main peak and the hole are still present, but several, much narrower and taller peaks appear on top of the wide distribution on the left of the hole. Although we do not have a clear interpretation for this observation, its appearance is clearly reminiscent of recoil effects (cf., e.g., Dopplerons [2]).

Finally, it is interesting to compare the predictions of the model [18] that includes random collisions and an arbitrary momentum distribution for the atoms—which can evolve during the interaction—and the original one [8] with the addition of the exponential relaxation [20]. In order to allow for a meaningful
comparison, the damping constant $\gamma$ has been fixed so as to correspond to the decay rate of correlations due to the collisions (i.e. $\gamma = 1/t_c$). In figure 5 one can see that the model with collisions (curves with no symbols) shows a much lower output intensity accompanied by a smaller value of the coherence parameter as well, when compared to the results of the model with exponential damping (curves with symbols). This is not too surprising, since collisions are continuously destroying the system’s self-organization. It is therefore reasonable to conclude that the predictions obtained on the basis of a simple exponential relaxation for the external degrees of freedom largely overestimate the effectiveness of the process. At the same time, it is interesting to notice that the transient evolution of the system is much shorter in our model. This is also understandable, since noise provides a *scrambling* mechanism that allows for faster reorganization of the atoms, starting from a generic initial condition, towards a somewhat ordered structure. However, the momentum distribution predicted by this model is very different from the typical shape shown figure 4. Thus, one may harbour some doubts about the validity of the approximate model which uses a damping term to describe the effect of collisions.

In conclusion, the introduction of collisions in the CARL model allows one to follow more closely the evolution of an atomic sample in interaction with a (strong) pump field. The presence of a thermal bath lets the atoms follow more realistic dynamical evolutions and arrange their velocity according to the interaction. As a consequence, total disappearance of the density grating occurs, which had been credited as the fundamental mechanism responsible for gain in the counter-propagating wave. Instead, a grating is observed in the *phase* of the atomic polarization—capable of surviving the perturbing effect of collisions thanks to its fast relaxation constant—and which is the likely source of the instability. An additional grating, in the population variable, is observed mainly as a consequence
of the interaction, while the polarization amplitude is, at most, weakly modulated; it is then concluded that the polarization grating (modulus and phase) which has been proposed as a possible alternative mechanism for the instability is not the true source term. Finally, it was remarked how the consideration of an exponential relaxation for the external atomic degrees of freedom overestimates the strength of the interaction and gives a wrong description of the momentum distribution.

Acknowledgments

We warmly thank L.M. Narducci and Z. Ye for fruitful discussions. M.P. is grateful to R. Kaiser for advice concerning the behaviour of cold atomic samples and G.L.L. to J.R. Tredicce for continuing exchanges on this problem. AP has done part of this work as Chercheur Associé CNRS.

References


The collective interaction between radiation and atoms