Quantum mechanical counterpart of nonlinear optics

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I. INTRODUCTION

A single atom trapped in a harmonic potential turns out to be a very well defined object for studying fundamental phenomena of quantum dynamics. Since the first realization of such a system in an ion trap by Neuhauser et al. [1], the subject has stimulated much experimental and theoretical work. As has been shown by Blockley et al. [2], the laser-assisted coupling between the internal and external degrees of freedom of a trapped atom can be described, under appropriate conditions, by a Jaynes–Cummings model. This allows one to study phenomena we are familiar with from cavity QED, such as the micromaser dynamics [3], in the vibronic motion of a trapped atom [1]. Eventually, several proposals have been published for preparing nonclassical states, such as squeezed states [4] and motional number states [5], and successful experiments have been performed [6,7].

The dynamics of a trapped atom, however, not only allows one to reproduce effects of cavity QED in the quantized motion. When the spatial extension of the atomic wave-function representing the center-of-mass motion is no longer small compared with the driving laser wavelength, nonlinear effects emerge that have no counterpart in standard nonlinear optics. It has been shown by Vogel and de Matos Filho that the atom may undergo a vibronic coupling which is very well described by a nonlinear, multiquantum Jaynes–Cummings model [8]. Meanwhile this prediction has been confirmed experimentally [9] and modifications due to micromotion have been studied [10]. The nonlinearities in this model allow to prepare exciting motional quantum states, such as quantum superpositions of both coherent [11] and squeezed states [12], nonlinear coherent states [13,14], pair coherent states [15] and pair cat-states [16]. Measurement techniques for the full diagnostics of motional quantum states have been proposed [17] and realized [18].

These outstanding feasibilities render it possible to rise new types of questions. The nonlinear Jaynes–Cummings model has introduced new kinds of nonlinearities that substantially modify phenomena we are familiar with from nonlinear optics, such as multiphoton absorption and emission. In nonlinear optics, however, other interactions are known which leave the electronic transitions of the nonlinear medium almost unchanged. Examples are the Kerr nonlinearity, parametric interactions and several types of nonlinear wave-mixings. The question appears as to whether it is possible to realize such phenomena in the motional dynamics of a single atom, where the trap potential replaces a cavity used in nonlinear optics.

In the present contribution we propose Raman-type excitations for inducing various kinds of nonlinear interactions in the quantized motion of a trapped atom. We consider the quantum mechanical counterpart of nonlinear optical effects that do not influence the electronic degrees of freedom of the atomic medium. We show that even a single degree of freedom of the atomic center-of-mass motion can be driven in a strongly nonlinear manner. Surprising phenomena are caused by the interference effects of the atomic wave function with the driving light waves. They induce a nonlinear partitioning of the phase space, the action of the time evolution being different in neighboring phase-space zones. This partitioning may be used for the generation of nonclassical effects like amplitude squeezing and quantum interferences.

The paper is organized as follows. In Sec. II the basic model for the Raman-induced motional dynamics is introduced and the effective Hamiltonian for the nonlinear motional interactions is derived. Section III is devoted to the nonlinear phase-space partitioning together with the illustration of its effects in simple examples of motional dynamics. A summary and some conclusions are given in Sec. IV.

II. RAMAN-INDUCED MOTIONAL DYNAMICS

Let us consider an atom harmonically bound in a trap. In general the atom oscillates in the three principal axes of the trap with frequencies \( \nu_i \) \((i = 1, 2, 3)\). The trapped atom is driven in a Raman configuration with two classical laser fields of frequencies \( \omega_L \) and \( \omega_L + \Delta \) \((\Delta \ll \omega_L)\), which are off-resonant with respect to the electronic tran-
by two off-resonant laser fields as coupling (in optical rotating-wave approximation) reads atom in a well-controlled manner. It is possible to affect the motional quantum state of the atom by choosing laser-beam geometry and laser detuning \( \Delta \), whereas other electronic states (broken lines) are far off-resonant. The beat frequency \( \Delta \) can be tuned on \( \omega_L \) and \( \omega_{21} \), respectively. Other electronic states (broken lines) remain a coupling of all vibrational modes. In this case the interaction Hamiltonian (in the interaction picture) is of the form \( H_{\text{int}}(\theta) \) of the laser waves, a laser-assisted coupling of the three motional degrees of freedom \( (x_1, x_2, x_3) \). Since the wave-vector difference \( k \) is determined by the laser-beam geometry, the coupling of the motional degrees of freedom can be designed to include one, two, or three directions.

To consider these couplings in more detail, we assume that the vibrational frequencies are well resolved by the Raman excitation, so that we may introduce a vibrational rotating-wave approximation. Choosing the laser beat frequency to be a multiple of the three vibrational frequencies, \( \Delta = s_1 \nu_1 + s_2 \nu_2 + s_3 \nu_3 \) \( (s_{1,2} = 0, \pm 1, \pm 2, \ldots) \), one obtains a coupling of all vibrational modes \( \nu_1, \nu_2, \nu_3 \). In this case the interaction Hamiltonian (in the interaction picture) is of the form \( H_{\text{int}}(\theta) \)

\[
\hat{H}_{\text{int}} = \frac{1}{2} \hbar \Omega \sum_{n=-\infty}^{\infty} \hat{g}_{n-s_1}(\hat{a}^\dagger_1, \hat{a}; \eta_1) \hat{g}_{n-s_2}(\hat{a}^\dagger_2, \hat{a}; \eta_2) \hat{g}_n(\hat{a}^\dagger_3, \hat{a}; \eta_3)
\]

\[+ H.c. \]

and the operator-valued functions \( \hat{g}_k(\hat{a}^\dagger, \hat{a}; \eta) \) are given by

\[
\hat{g}_k(\hat{a}^\dagger, \hat{a}; \eta) = \begin{cases} 
(i \eta \hat{a}^\dagger)^{|k|} \hat{f}_k(\hat{n}; \eta) & \text{if } k \geq 0 \\
\hat{f}_k(\hat{n}; \eta) (i \eta \hat{a})^{|k|} & \text{if } k < 0
\end{cases}
\]

The Hermitian operator functions \( \hat{f}_k(\hat{n}; \eta) \) depend solely on the number of vibrational quanta \( \hat{n} = \hat{a}^\dagger \hat{a} \) and read (in normally ordered form) as

\[
\hat{f}_k(\hat{n}; \eta) = e^{-\eta^2/2} \sum_{l=0}^{\infty} \frac{(-1)^l \eta^{2l}}{l!(l+k)!} \hat{a}^l \hat{a}^\dagger
\]

From Eqs. (3) and (4) it is seen, that for decreasing Lamb–Dicke parameter only the coupling with \( k = 0 \) survives. Therefore, by varying the geometry of the laser-beam propagation one can vary the Lamb–Dicke parameters in order to change the Hamiltonian from a coupling of only one, two, or three vibrational modes.

It is seen from Eqs. (3), (4) that the Hamiltonian describes a motional dynamics with the following basic effects. First, there appear combinations of different powers of the motional operators \( \hat{a}, \hat{a}^\dagger \). Interactions of this type represent the quantum mechanical counterpart of Lamb–Dicke parameters of the vibration in these directions. After disentangling the resulting exponential operator function, the Hamiltonian \( \hat{H}_{\text{int}}(\theta) \) may be expanded in a power series as

\[
\hat{H}_{\text{int}}(t) = \frac{1}{2} \hbar \Omega e^{i\Delta t} e^{-(\eta_1^2 + \eta_2^2 + \eta_3^2)/2} \sum_{m m' n n'} \sum_{l l'} (i \eta_{m m'} (i \eta_{n n'})^{l l'}) \frac{i}{m! n! n'! l! l'} \hat{a}^m \hat{a}_{m'}^n \hat{a}^l \hat{a}_{l'}^n + H.c.
\]
wave-mixing effects in nonlinear optics. Second, via the functions \( f_k(\hat{n}; \eta) \) the couplings depend in a nonlinear manner on the excitations of the modes. This results from the interference of the atomic (center-of-mass) wave functions and the beat node of the laser waves, which is a typical effect of quantized atomic motion.

III. NONLINEAR PHASE-SPACE PARTITIONING

To get some insight in these effects, we first consider the one-dimensional dynamics, where only the motion in \( x_1 \)-direction is affected by the lasers \((\gamma_2 = \gamma_3 = 0)\). This requires a geometry of laser propagations with vanishing projections of the difference wave-vector \( k \) on the axes \( x_2 \) and \( x_3 \). In this case the Hamiltonian simplifies as

\[
\hat{H}_{\text{int}} = \frac{1}{2} \hbar \Omega f_k(\hat{n}; \eta) (i\eta \hat{a})^k + \text{H.c.}, \tag{7}
\]

where we assumed a laser detuning of \( \Delta = k\nu_1 \) \((k \geq 0)\) and we have omitted the indices of the \( x_1 \) direction. Interactions of this type may be considered as nonlinear mode couplings of one (weakly excited) quantized mode with (strongly excited) classical modes. Such approximations are frequently used in quantum optics. Experiments of the type proposed here would allow to realize these couplings almost perfectly and to study the additional (excitation-dependent) nonlinearities.

For example, let us consider the one-quantum resonance \( (\Delta = \nu_1) \) in more detail. In this case the structure of the unitary time-evolution operator obtained from the Hamiltonian \( (8) \) shows some formal resemblance to a nonlinearly modified coherent “displacement” operator \([2]\),

\[
\hat{U}_{\text{int}}(t) = \hat{D} \left[ \frac{-\eta \Omega_t}{2} \hat{f}_1(\hat{n}; \eta) \right] = \exp \left[ \frac{-\eta \Omega t}{2} \hat{a}^3 \hat{f}_1(\hat{n}; \eta) + \frac{\eta \Omega t}{2} \hat{f}_1(\hat{n}; \eta) \hat{a} \right]. \tag{8}
\]

For small values of the Lamb–Dicke parameter, \( \eta \ll 1 \), according to Eq. (8) the operator \( (8) \) may be replaced by the usual displacement operator \( \hat{D}(-\eta \Omega t/2) \).

The nonlinear dependence of the “displacement” operator \( (8) \) on the mean number of vibrational quanta leads to effects of a new type. For a first insight we may replace the number operator by its eigenvalue. We arrive at the c-number function \( f_1(n; \eta) = \langle n | \hat{f}_1(\hat{n}; \eta) | n \rangle \), which reads as

\[
f_1(n; \eta) = e^{-n^2/2} L_n^{(1)}(\eta^2), \tag{9}
\]

with \( L_n^{(k)}(x) \) being Laguerre polynomials. To consider the action of the nonlinear displacement in phase space, it is advantageous to introduce the (complex) phase-space amplitude \( \alpha \) by setting \( n = |\alpha|^2 \). The resulting function \( f_1(|\alpha|^2; \eta) \) has zeros and changes its sign for certain values of \( |\alpha| \). Consequently, the direction of the displacement can be reversed, depending on the amplitude of the quantum state in phase space. That is, the phase space is effectively partitioned in zones. The action of the displacement in adjacent zones differs in the fact that the directions of displacements are opposite to each other, along an axis which is controlled by the phase difference of the lasers. These phase-space zones are separated by the circles on which the coupling function \( f_1(|\alpha|^2; \eta) \) changes its sign. This nonlinear partitioning of the phase space leads to striking consequences with respect to the evolution of the quantum state.

Let us consider the evolution of a coherent state that is initially located on the boundary between two such phase-space zones. Inside the corresponding circle the coupling \( f_1(|\alpha|^2; \eta) \) is positive and outside it is negative. Due to this fact the nonlinear “displacement” operator tends to split the coherent state as shown in Fig. 2. For rather short times the state can exhibit a significant reduction of phase fluctuations. In the further course of time the states is splitted into well separated substates. This leads to a coherent superposition of two quantum states, accompanied by quantum-interference effects. The displacement of each substate is limited by the boundaries between the phase-space zones, where the strength of displacement becomes negligible. The result

![Fig. 2. Time evolution of a coherent state that is initially placed on the boundary between two phase-space zones with opposite displacement directions (chosen along the real axis). The dimensionless times \( \eta(\Omega t) \) are given by 0 (a), 2.5 (b), 5 (c), and 15 (d); \( \eta = 0.25 \). The contours represent the \( Q \) functions of the motional quantum states.](image-url)
is a squeezing of each substate onto the corresponding circle partitioning the phase space.

This effect can be used to generate quantum states exhibiting strong amplitude squeezing. Let us consider the nonlinear displacement of a coherent state that is initially located within a single phase-space zone. As expected, the state is displaced in a well defined direction in phase space until it is squeezed onto the next circle separating two zones. The result consists in a strongly amplitude-squeezed state \( |\eta\rangle \) with a non-vanishing coherent amplitude as shown in Fig. 3. It is worth noting that in its further evolution this quantum state does not approach the state is displaced in a well defined direction in phase space where the change in sign of the coupling.

For \( k = 0 \) the time-evolution operator agrees with the squeez operator. This limiting case has been realized experimentally \( [22] \). In the more general case of larger Lamb–Dicke parameters, a rather complex dynamics appears. The interpretation of all of its features needs some further research.

For studying a quantized version of the parametric interaction, the coupling of two degrees of freedom is needed. Consider a laser-beam geometry with the projection of the difference wave-vector \( k \) on the \( x_3 \)-axis being zero, so that \( \eta_3 = 0 \). The dynamics couples the motion in \( x_1 \) and \( x_2 \) directions. For example, a detuning of \( \Delta = 2\eta_1 - \eta_2 \) \( (s_1 = 2, s_2 = -1) \) reduces the interaction Hamiltonian \( [4] \) to

\[
\hat{H}_{\text{int}} = -\frac{i}{2} \hbar f_0^2 \Omega \frac{\hat{a}_1^\dagger \hat{a}_2^\dagger f_{1}(\hat{n}_1; \eta_1) \hat{a}_1^\dagger \hat{a}_2 f_{1}(\hat{n}_2; \eta_2)}{\Omega} + H.c.,
\]

representing a nonlinear generalization of the parametric interaction. For small Lamb–Dicke parameters, \( \eta_{1,2} \ll 1 \), this interaction simplifies to

\[
\hat{H}_{\text{int}} = -\frac{i}{2} \hbar f_0^2 \Omega \frac{\hat{a}_1^\dagger \hat{a}_2^\dagger + H.c.}{\Omega},
\]

which is the standard form of the parametric coupling. Beyond the Lamb–Dicke regime the interaction includes nonlinearities of the type considered above, which now appear in both motional degrees of freedom. Consequently, the nonlinear phase-space partitioning effects considered above will be of relevance for each degree of freedom involved in the Raman-induced motional dynamics.
IV. SUMMARY AND CONCLUSIONS

In conclusion we have shown, that a Raman-type laser excitation allows to induce nonlinear interactions of motional degrees of freedom of a trapped atom, which are closely related to phenomena of nonlinear optics that do not change the electronic quantum states of the medium. The number of coupled modes can be easily controlled by the laser beam geometry. Standard effects can be realized, including coherent displacements, Kerr nonlinearities, and parametric mode couplings. In the laser-assisted motional dynamics additional nonlinearities emerge, which are caused by the interference between the light waves and the wave function representing the atomic center-of-mass motion.

An important consequence of these nonlinearities consists in a partitioning of the motional phase space, which is caused by an oscillatory behavior of the motional interactions as a function of the phase-space amplitude. In neighboring phase-space zones the actions of the time evolution appear to be significantly different from each other. For example, in two adjacent zones a nonlinearly modified "displacement" operator acts in opposite directions. Consequently, a quantum state whose initial location is on the boundary between two zones will be split in two substates, which eventually gives rise to quantum interferences. Moreover, the partitioning allows to generate strongly amplitude-squeezed motional states. Eventually, in the case of a generalized Kerr nonlinearity the phase-space partitioning may lead to pronounced deformations of the initial state, which are caused by opposite phase shifts appearing in adjacent phase-space zones.

The phase-space partitioning, although illustrated in this paper for the motional dynamics in one dimension, is a universal feature of the interference between the Raman beat node and the wave function describing the center-of-mass motion of the atom. When two or three dimensions are involved in the Raman-induced dynamics, the partitioning effects appear in the phase space of each motional degree of freedom. Consequently, the coupling between different motional modes will be strongly influenced by the interplay of these nonlinear effects. In general the dynamics will sensitively depend on the initial conditions. Besides the feasibility of realizing phenomena well known from nonlinear optics in the motion of a trapped atom, this opens novel possibilities for studying nonlinear phenomena in a well-defined quantum system.

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a consequence of the fact that the operator-valued function \( f_1 \) does not commute with the annihilation and creation operators. However, its action shows some resemblance to a nonlinear displacement which depends on the amplitude of the quantum state in phase space.

[22] Note that a hint on such an amplitude-squeezing effect has already been found experimentally, cf. footnote 36 of Ref. [8].

[23] A different scheme to realize Kerr-type effects of atomic motion has been proposed by J.K. Breslin, C.A. Holmes, and G.J. Milburn, Phys. Rev. A, submitted.