Many-body aspects of coherent atom-molecule oscillations

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(Dated: February 18, 2003)

PACS numbers: 03.75.Kk, 67.40.-w, 32.80.Pj

We study the many-body effects on coherent atom-molecule oscillations by means of an effective quantum field theory that describes Feshbach-resonant interactions in Bose gases in terms of an atom-molecule Hamiltonian. We determine numerically the many-body corrections to the oscillation frequency for various densities of the atomic condensate. We also derive an analytic expression that approximately describes both the density and magnetic-field dependence of this frequency near the resonance. We find excellent agreement with experiment.

Introduction — One of the most remarkable applications of Feshbach resonances in doubly spin-polarized alkali gases [1, 2, 3] is the observation of coherent atom-molecule oscillations [4]. In this last experiment Donley et al. used the Feshbach resonance at \( B_0 \approx 154.9 \) Gauss in the \(|f = 2; m_f = -2\) state of \(^{85}\)Rb to perform a Ramsey-type experiment, consisting of two short pulses in the magnetic field towards resonance separated by a longer evolution time. As a function of this evolution time an oscillation in the number of condensate atoms was observed. Over the investigated range of magnetic field during the evolution time, the frequency of this oscillation agreed exactly with the molecular binding energy found from a two-body coupled-channels calculation [5], indicating coherence between atoms and molecules.

Very recently, Claussen et al. have performed a similar series of measurements over a larger range of magnetic fields [6]. It was found that close to resonance the frequency of the oscillation deviates from the two-body molecular binding energy, which indicates that many-body effects play an important role in this regime. It is the main purpose of this Letter to present the theory that explains this deviation quantitatively.

The first mean-field theory for Feshbach-resonant interactions in Bose-Einstein condensed gases is due to Drummond et al. [7] and Timmermans et al. [8] and introduces the physical picture of an interacting atomic condensate coupled to a noninteracting molecular condensate. Although this theory contains the correct resonant scattering amplitude for the atoms, it does not contain the correct molecular binding energy. By studying the fluctuations around this mean-field theory within the Hartree-Fock-Bogoliubov approximation, it is possible to also incorporate the correct binding energy [7, 8, 9]. This comes about because the latter approach explicitly contains a so-called anomalous density, or pairing field, for the atoms which, after elimination, leads to a shift in the coupling constants and the molecular binding energy. Unfortunately, however, the elimination does not lead to a proper renormalization of all the coupling constants. In particular the interaction between condensate atoms and non-condensate atoms, which is ultimately responsible for the many-body corrections to the molecular binding energy that are of interest to us here, is not correctly described within the Hartree-Fock-Bogoliubov approximation.

Relying on the anomalous density for the description of the molecular properties also makes the theory inapplicable above the critical temperature for Bose-Einstein condensation, since the anomalous density is proportional to the atomic condensate density. To overcome all these problems, it is convenient to formulate an effective quantum field theory that incorporates the exact two-body physics not at the mean-field level but at the quantum level and is hence applicable both above and below the critical temperature. We have recently derived such an effective quantum field theory by starting from the microscopic hamiltonian for an atomic gas with a Feshbach resonance and explicitly summing all the ladder diagrams [10]. Here, we apply the mean-field theory for the Bose-Einstein condensed phase of the gas that results from this effective quantum field theory, to the study of the recent experiments by Claussen et al. [6].

Atom-molecule coherence — The mean-field equations consist of coupled equations for the macroscopic wave function \( \psi_a(x, t) \), describing the atomic Bose-Einstein condensate, and the macroscopic wave function \( \psi_m(x, t) \) that describes the condensate of bare molecules, i.e., with the appropriate bound-state wave function in the closed channel potential of the Feshbach problem. The mean-field equations for the coupled atom-molecule system are ultimately given by

\[
\begin{align*}
\frac{i\hbar}{\partial t} \psi_a(x, t) &= \left[ -\frac{\hbar^2 \nabla^2}{2m} + T_{bg}^{2B} |\psi_a(x, t)|^2 \right] \psi_a(x, t) + 2g \psi_a^* (x, t) \psi_m (x, t), \\
\frac{i\hbar}{\partial t} \psi_m (x, t) &= \left[ -\frac{\hbar^2 \nabla^2}{4m} + \delta(B(t)) \right] \psi_m (x, t) + g \psi_a^2 (x, t) \\
&- g^2 n^{3/2} \frac{2\pi \hbar^2}{m} \sqrt{\frac{i\hbar}{\partial t} + \frac{\hbar^2 \nabla^2}{4m} - 2\hbar \Sigma^{HF}} \psi_m (x, t),
\end{align*}
\]

where \( T_{bg}^{2B} = 4\pi a_{bg} \hbar^2 / m \) is the off-resonant two-body
T(transition) matrix with $a_{bg}$ the off-resonant background scattering length and $m$ the mass of one atom. The atom-molecule coupling $g$ is found from experiment by adiabatically eliminating the molecular wave function and using the fact that the resulting magnetic-field dependent scattering length of the atoms must be equal to
\[ a(B) = a_{bg} \left( 1 - \frac{\Delta B}{B - B_0} \right), \] (2)
with $\Delta B$ and $B_0$ the experimental width and position of the resonance, respectively. This procedure results in $g = \hbar/\sqrt{2\pi a_{bg}} \Delta B \Delta \mu/m$, where we have made use of the fact that the detuning of the bare molecular state is given by $\delta(B) = \Delta \mu(B - B_0)$ with $\Delta \mu \approx -2.2 \mu_B$ for $^{85}\text{Rb}$ and $\mu_B$ the Bohr magneton.

The mean-field equation for the molecular condensate contains a fractional derivative, corresponding to the retarded self energy of the molecules. In momentum and frequency space, this self energy reads $\hbar \Sigma^{(+)}(k, \omega) = -(g^2 m^{3/2}/2\pi\hbar^2)i\sqrt{\hbar \omega - \hbar^2 k^2/4m - 2\hbar \Sigma^{HF}}$. The square-root behavior is a result of the Wigner threshold law for the decay of a molecule with total energy $\hbar \omega$ and center-of-mass momentum $\hbar k$ into the two-atom continuum. Due to the mean-field interaction of the noncondensed atoms with the condensate, the decaying molecule has to overcome a mean-field barrier given by $2\hbar \Sigma^{HF}$. Here, $\hbar \Sigma^{HF}$ denotes the Hartree-Fock self energy for the noncondensed atoms. Neglecting the momentum dependence of this self energy, it is given by
\[ \hbar \Sigma^{HF} = 2n_a \left( T_{bg}^{2B} + \frac{2g^2}{\hbar \Sigma^{HF} + \mu - \delta(B) - g^2 m^{3/2}/2\pi\hbar \sqrt{\hbar \Sigma^{HF} - \mu}} \right), \] (3)
where $n_a = |\psi_a|^2$ is the density of the atomic condensate, $\mu$ its chemical potential, and we have used that a collision between a condensed atom and a noncondensed atom has a mean-field shift of $\mu + \hbar \Sigma^{HF}$. Far from resonance the energy-dependence of the interactions can be safely ignored and the Hartree-Fock self energy becomes equal to $8\pi a(B)\hbar^2 n_a/m$, as expected.

The molecular binding energy is given by the pole of the molecular propagator at zero momentum, which from Eq. (1) is seen to be given by
\[ G_m^{(+)}(0, \omega) = \frac{\hbar}{\hbar \omega + i0 - \delta(B) + g^2 m^{3/2}/2\pi\hbar \sqrt{\hbar \omega - \hbar \Sigma^{HF}}}. \] (4)

In the limit of vanishing condensate density $n_a$ and for negative detuning, it has a pole at
\[ \epsilon_m(B) = \delta(B) + \frac{g^4 m^3}{8\pi^2 \hbar^6} \left[ \sqrt{1 - \frac{16\pi^2 \hbar^6}{g^4 m^3} \delta(B)} - 1 \right]. \] (5)

Close to the resonance it thus follows that $\epsilon_m(B) = -\hbar^2/\left[ma(B)^2\right]$, which is the correct molecular binding energy in vacuum [3]. Note that in the absence of the molecular self energy the binding energy would be equal to the detuning, which is incorrect close to resonance. Moreover, the residue of the pole is given by
\[ Z(B) = \left[ 1 - \frac{\partial \Sigma_m^{(+)}(0, \omega)}{\partial \omega} \right]^{-1} \]
\[ = \left[ 1 + \frac{g^2 m^{3/2}}{4\pi\hbar^3 \sqrt{\epsilon_m(B)}} \right]^{-1} \] (6)
and always smaller than one. Physically, the latter can be understood from the fact that the dressed molecular bound state near the Feshbach resonance is given by
\[ |\chi_m; \text{dressed}\rangle = \sqrt{Z(B)}|\chi_m; \text{bare}\rangle + \int \frac{dk}{(2\pi)^3} C(k)|-k; \text{open}\rangle, \] (7)
where the coefficient $C(k)$ denotes the amplitude of the dressed molecular state to be in the open channel of the Feshbach problem and the two atoms having momenta $k$ and $-k$, respectively. They are normalized as $\int dk|C(k)|^2/(2\pi)^3 = 1 - Z(B)$. The dressed molecular state therefore only contains with an amplitude $\sqrt{Z(B)}$ the bare molecular state $|\chi_m; \text{bare}\rangle$. Close to resonance we have that $Z(B) \ll 1$, whereas it approaches one far off resonance. With respect to this remark it is important to note that the result of the Hartree-Fock-Bogoliubov theory for the density of the molecular condensate should be multiplied by a factor $1/Z(B) \gg 1$ to obtain the density of real dressed molecules, since in this theory always the density of bare molecules is calculated [12, 13].

**Collective modes** — To study the many-body effects on the frequency of the coherent atom-molecule oscillations it is important to realize that these oscillations are in fact a collective mode where the atomic condensate density oscillates out-of-phase with the molecular condensate density. It is thus worthwhile to study the collective modes of the mean-field equations in Eq. (1) and look for solutions of the form
\[ \psi_m(x, t) = \left[ \psi_m + u_k e^{i\omega t + i\mathbf{k}\cdot\mathbf{x}} + \psi^{*}_m e^{i\omega t - i\mathbf{k}\cdot\mathbf{x}} \right] e^{-i\mu t/\hbar}, \]
\[ \psi_n(x, t) = \left[ \psi_n + u_k e^{i\omega t + i\mathbf{k}\cdot\mathbf{x}} + \psi^{*}_n e^{i\omega t - i\mathbf{k}\cdot\mathbf{x}} \right] e^{-i\mu t/\hbar}. \] (8)

After substitution into the mean-field equations in Eq. (1) the eigenmodes are found by diagonalizing the resulting $4 \times 4$ matrix. This yields a dispersion relation with two branches, one corresponding to the gapless Bogoliubov modes and one that corresponds to the atom-molecule oscillations. The zero-momentum part of the latter corresponds to the experimentally observed frequency of the coherent atom-molecule oscillations. Note that for this calculation the evaluation of the molecular
sion of the lower branch obeys \( \bar{\omega} \), actions that reduce the scattering amplitude. The dispersion, as expected. At higher momenta the dispersion is negative due to the fact that we are dealing with a metastable situation. For negative detuning the lower branch obeys \( \bar{\omega} \), whereas both quantities individually show a substantial cancellation, however, also the wave-function renormalization factor \( Z \) is almost independent of density and equal to \( \epsilon_m(B) \). It is interesting to note that only this difference is independent of density, whereas both quantities individually show a substantial mean-field shift of about \( 2\hbar \Sigma_{HF} \). As a result of the cancellation, however, also the wave-function renormalization factor \( Z \) is almost density independent and equal to \( Z(B) \). Expressing our mean-field equations in Eq. (10) in terms of the condensate wave function for the dressed molecules by replacing \( \psi_m(x,t) \) by \( \sqrt{Z(B)}\psi_m(x,t) \), we see that the coupling between the atomic condensate and the dressed molecular condensate is reduced by a factor of \( \sqrt{Z(B)} \ll 1 \). We thus expect the frequency to obey

\[
\hbar \omega_3 \simeq \sqrt{16Z(B)g^2n_a + (\epsilon_m(B))^2} .
\]

This analytic expression indeed turns out to give a first approximation to the deviation of the frequency from the two-body result.

In order to confront our results with the experimental data we have to realize that the experiments are performed in a magnetic trap. Taking only the ground-states into account for both the atomic and the molecular condensates, this implies effectively that the atom-molecule coupling \( g \) is reduced by an overlap integral. Hence we define the effective homogeneous condensate density by

In Fig. \( \Box \) we show the dispersion relations found by means of the above procedure, for a fixed atomic condensate density of \( n_a = 2 \times 10^{12} \text{ cm}^{-3} \), at a magnetic field of \( B = 156 \text{ G} \). Physically, the upper branch corresponds to the phonon-like excitations in the atomic condensate and the lower branch to coherent atom-molecule oscillations. The dashed line denotes the Bogoliubov dispersion for the scattering length \( a(B) \). At low momenta the phonon branch corresponds with the Bogoliubov dispersion, as expected. At higher momenta the dispersion starts to deviate from the Bogoliubov result, due to the energy and momentum dependence of the resonant interactions that reduce the scattering amplitude. The dispersion of the lower branch obeys \( \hbar \omega_3 \simeq -\hbar \omega_3 + \hbar^2 k^2/4m \), where \( \hbar \omega_3 \) is the Josephson frequency that is observed by Claussen et al. in their Ramsey experiment. This dispersion is negative due to the fact that we are dealing with a metastable situation. For negative detuning the true ground state contains almost all atoms in the form of molecules.

In Fig. \( \Box \) the results are shown for the frequency of the atom-molecule oscillations as a function of the magnetic field, for three different densities. Clearly, for increasing density the frequency starts to deviate from the two-body result. In addition, we show in Fig. \( \Box \) the frequency of the atom-molecule oscillations relative to the two-body binding energy, as a function of the atomic condensate density. The calculation is performed in this case for several values of the magnetic field. Both the magnetic field and the atomic density dependence of the frequency can be understood as follows. We first observe that for the range of magnetic fields and atomic densities that are explored experimentally, the difference between the energy of the dressed molecular state and the threshold of the two-particle continuum is almost independent of density and equal to \( \epsilon_m(B) \). It is interesting to note that only this difference is independent of density, whereas both quantities individually show a substantial mean-field shift of about \( 2\hbar \Sigma_{HF} \). As a result of the cancellation, however, also the wave-function renormalization factor \( Z \) is almost density independent and equal to \( Z(B) \). Expressing our mean-field equations in Eq. (10) in terms of the condensate wave function for the dressed molecules by replacing \( \psi_m(x,t) \) by \( \sqrt{Z(B)}\psi_m(x,t) \), we see that the coupling between the atomic condensate and the dressed molecular condensate is reduced by a factor of \( \sqrt{Z(B)} \ll 1 \). We thus expect the frequency to obey

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means of $n_a = N_a \left[ \int dx \phi^2_a(x) \phi_m(x) \right]^2$, where $N_a$ denotes the number of condensed atoms and $\phi_a(x)$ and $\phi_m(x)$ denote the atomic and molecular ground state wave function, respectively. For the experiments of Claussen et al. this results in an effective density of $n_a \approx 2 \times 10^{12}$ cm$^{-3}$ Fig. 2 clearly shows an excellent agreement with the experimentally observed frequency for this density.

It is important to note that there are two hidden assumptions in the above comparison. First, we have used that the dressed molecules are trapped in the same external potential as the atoms. This is not obvious because the bare molecular state involved in the Feshbach resonance is high-field seeking and therefore not trapped. However, Eq. (6) shows that near resonance almost all the amplitude of the dressed molecule is in the low-field seeking open channel and its magnetic moment is therefore almost equal to twice the atomic magnetic moment. Second, we have determined the frequency of the coherent atom-molecule oscillations in equilibrium. In contrast, the observed oscillations in the number of condensed atoms is clearly a nonequilibrium phenomenon. This is, however, expected not to play an important role because the Ramsey-pulse sequence is performed on such a fast time scale that the response of the condensate wave function can be neglected.

Conclusions — With the linear-response calculation presented in this Letter we have obtained excellent agreement with the experimental results on the frequency of coherent atom-molecule oscillations. The next step is a more detailed understanding of other quantities that are of interest for the two-pulse experiments. This includes a quantitative study of the number of condensed atoms as a function of time that goes beyond the linear approximation discussed here. Experimentally an over-all decay of the number of condensed atoms is observed and also that the oscillations have a finite damping rate. Both effects increase as one approaches the resonance. We expect that an important contribution to these effects is due to the so-called rogue-dissociation process, which has its physical origin in the decay of a molecule into two noncondensed atoms that in the center-of-mass system have opposite momenta $\mathbf{k}$ and $-\mathbf{k}$, respectively. In equilibrium this process is always forbidden due to energy conservation, since the molecular state lies below the two-atom continuum threshold. However, in the nonequilibrium setting of the experiments of interest, it may occur due to the fact that the detuning is strongly time-dependent. In the mean-field equations in Eq. (11) it is the fractional derivative term corresponding to the self energy of the molecules that automatically incorporates the rogue-dissociation effect. A full numerical simulation of these equation is challenging due to the fact that we are essentially dealing with a term that is nonlocal in time. Nevertheless, work in this direction is in progress and will be reported in a future publication.

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13. See also E. Braaten, H.-W. Hammer and M. Kusunoki (cond-mat/0301489).
14. The full expression for the self energy of the molecules is given by $\Sigma^{(3)}(k, \omega) = -\left( q^2 m^{3/2} / 2\pi \hbar^2 \right) \sqrt{\pi} (1 + imn_a \sqrt{\omega^2 - \hbar^2 k^2 / 4m - 2\Sigma^{HF}})^{-1}$, where $z = \hbar \omega - \hbar^2 k^2 / 4m - 2\Sigma^{HF}$. Close to resonance only the square-root in the numerator is important. We will use this approximation throughout the paper.