Nonlinear Response Functions of Strongly Correlated Boson Fields: Bose-Einstein Condensates and Fractional Quantum Hall Systems

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The second order response functions and susceptibilities of finite temperature Bose-Einstein Condensates (BEC) in a one dimensional harmonic trap driven by an external field that couples to the particle density are calculated by solving the time-dependent Hartree-Fock-Bogoliubov (TDHFB) equations. These provide additional insight into BEC dynamics, beyond the linear response regime. The results also apply to electron liquids in the Fractional Quantum Hall Effect (FQHE) regime which can be mapped onto an effective boson system coupled to a Chern-Simons gauge field.

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I. INTRODUCTION

We have recently studied an externally driven, finite temperature Bose-Einstein condensates (BEC) described by the time-dependent Hartree-Fock-Bogoliubov (TDHFB) equations \cite{1}. A systematic procedure was outlined for solving these equations perturbatively in the applied external field, and position-dependent linear response functions and susceptibilities were calculated.

In this paper we extend this formalism to calculate the second order response function for the condensate, non-condensate density and non-condensate correlation. The linear response provides an adequate description of the system only for weak external perturbations. Otherwise, nonlinear effects contained in the higher order terms in the perturbation series may not be ignored. There are strong similarities between nonlinear optics and the dynamics of BEC owing to the interatomic interactions in Bose condensates. Previous work done in this area includes the demonstration of four wave mixing in zero temperature BEC using the Gross-Pitaevskii Equation (GPE) \cite{2}. In the present article we study nonlinear properties of BEC using the TDHFB framework.

Numerous dynamical theories exist for finite temperature BEC that takes into account higher order collision processes, such as the time-dependent Bogoliubov-de Gennes equations \cite{3}, the Hartree-Fock-Bogoliubov (HFB) theory \cite{4,5,6,7}, Quantum Kinetic Theory \cite{8,9,10,11,12,13}, and Stochastic methods \cite{14,15,16,17,18}. The TDHFB theory is a self-consistent theory of BEC in the collisionless regime that progresses logically from the Gross-Pitaevskii Equation by taking into account higher order correlations of noncondensate operators. Although TDHFB neglects higher order correlations included in the various quantum kinetic theories, the TDHFB equations are valid at very low temperatures near zero, even down to the zero temperature limit, and are far simpler than the kinetic equations which can only be solved using approximations such as ZNG. Another attractive feature of TDHFB from a purely pragmatic point of view is that the Fermionic version of the theory has already been well-developed in Nuclear Physics \cite{19}. We therefore work at the TDHFB level in this paper and our approach draws upon the analogy with the time-dependent Hartree-Fock (TDHF) formalism developed for nonlinear optical response of many electron systems \cite{20}.

In Section II we introduce the second order time and frequency domain response functions for an externally driven BEC. Numerical results are discussed in Sections III and IV for a condensate of 2000 atoms in a one dimensional harmonic trap. In Section V we show how this formalism may be applied for computing the second order response for a Fractional Quantum Hall system. In Section VI we conclude. Details of the derivation of the second order response functions are given in the Appendix.

II. NONLINEAR RESPONSE FUNCTION OF EXTERNALLY DRIVEN BEC

We adopt the notation of Ref. \cite{1} throughout the paper. The Hamiltonian describing the system of an externally driven, trapped atomic BEC is given by:

\begin{equation}
\hat{H} = \hat{H}_0 + \hat{H}'(t),
\end{equation}
where
\[ H_0 = \sum_{ij} H^{sp}_{ij} \hat{a}_i^\dagger \hat{a}_j + \frac{1}{2} \sum_{ijkl} V_{ijkl} \hat{a}_i^\dagger \hat{a}_j \hat{a}_k \hat{a}_l, \]  
(2)
and
\[ H'(t) = \eta \sum_{ij} E_{ij}(t) \hat{a}_i^\dagger \hat{a}_j \]  
(3)
with
\[ E_{ij}(t) = \int d^3r \phi_i^*(r) V_f(r) \phi_j(r) \hat{a}_i^\dagger \hat{a}_j. \]  
(4)

The boson operators \( \hat{a}_i^\dagger \) (\( \hat{a}_i \)) create (annihilate) a particle from a basis state with wave functions \( \phi_i(r) \). The single particle Hamiltonian \( H^{sp} \) in Eq. (2) is diagonal if the basis state \( \phi_i(r) \) is chosen to be the eigenstates of the trap, while the symmetrized interaction matrix elements,

\[ V_{ijkl} = \int d^3r d^3r' \phi_i^*(r) \phi_j^*(r') V(r - r') \phi_k(r) \phi_l(r), \]  
(5)
describe the collision between the atoms, with \( V(r - r') \) being a general interatomic potential. \( H'(t) \) describes the effect of a general external force \( V_f(r, t) \) on the condensate that mimics the mechanical force applied experimentally such as shaking of the trap [25, 30].

The dynamics of the system is calculated by solving the time-dependent Hartree-Fock-Bogoliubov (TDHFB) equations for the condensate mean field, \( z_i = \langle \hat{a}_i \rangle \), the non-condensate density \( \rho_{ij} = \langle \hat{a}_i^\dagger \hat{a}_j \rangle - \langle \hat{a}_i^\dagger \rangle \langle \hat{a}_j \rangle \), and the non-condensate correlations \( \kappa_{ij} = \langle \hat{a}_i \hat{a}_j \rangle - \langle \hat{a}_i \rangle \langle \hat{a}_j \rangle \). These are presented in Appendix A. These nonlinear coupled equations are solved by an order by order expansion of the variables \( z_i \), \( \rho_{ij} \) and \( \kappa_{ij} \); at each order, the resulting equations to be solved become linear [1]. It is found that the sequence of linear equations to be solved has the general form:

\[ i\hbar \frac{d\tilde{\psi}^{(n)}(t)}{dt} = \mathcal{L}^{(n)}(t) \tilde{\psi}^{(n)}(t) + \lambda^{(n)}(t), \]  
(6)
where we have denoted the set of \( n \)th order variables \( z_i^{(n)} \), \( \rho_{ij}^{(n)} \), and \( \kappa_{ij}^{(n)} \) as a \( 2N(2N + 1) \times 1 \) column vector in the Liouville space notation where \( N \) is the number of basis states used, \( \tilde{\psi}^{(n)} = [z^{(n)}, z^{(n)*}, \rho^{(n)}, \kappa^{(n)}, \rho^{(n)*}, \kappa^{(n)*}]^T \), and the \( 2N(2N + 1) \times 2N(2N + 1) \) matrix \( \mathcal{L}^{(n)} \) is the \( n \)th order Liouville operator obtained from the TDHFB equations [1]. The matrices \( \mathcal{L}^{(n)} \) for all orders \( n > 0 \) are identical i.e. \( \mathcal{L}^{(1)} = \mathcal{L}^{(2)} = \cdots = \mathcal{L}^{(n)} \) so that only the matrices \( \mathcal{L}^{(0)} \) and \( \mathcal{L}^{(1)} \) are required to be calculated. \( \mathcal{L}^{(0)} \) and \( \mathcal{L}^{(1)} \) are presented in Appendix B. The formal solution to Eq. (6) is:

\[ \tilde{\psi}^{(n)}(t) = \frac{1}{i\hbar} \int_0^t \exp \left[ -\frac{i}{\hbar} \mathcal{L}^{(n)}(t - t') \right] \lambda^{(n)}(t') dt'. \]  
(7)
In the frequency domain, the solution to Eq. (6) takes the form:

\[ \tilde{\psi}^{(n)}(\omega) = \frac{1}{\omega - \mathcal{L}^{(n)}} \lambda^{(n)}(\omega), \]  
(8)
where \( \tilde{\psi}^{(n)}(\omega) \) and \( \lambda^{(n)}(\omega) \) are the Fourier transforms of \( \tilde{\psi}^{(n)}(t) \) and \( \lambda^{(n)}(t) \) respectively.

For the zero’th order \( (n = 0) \), we obtain the time-independent HFB equations (TIHFB)

\[ \mathcal{L}^{(0)} \tilde{\psi}^{(0)}(t) = 0, \]  
(9)
while for the first order \( (n = 1) \), the equation solved is Eq. (6) with \( \lambda^{(1)}(t) \) being a \( 2N(2N + 1) \times 1 \) vector \( \zeta(t) \) calculated in Ref. [1]; \( \zeta(t) \) is also presented in Appendix B.

Once the \( n \)th order solution to TDHFB is found, we can proceed to define the \( n \)th order response functions. The physical significance of the response functions become more transparent when the \( n \)th order solutions \( \alpha^{(n)} \) where \( \alpha \)
is one of the variables $z$, $\rho$ or $\kappa$ are expressed in real space. We therefore introduce the position dependent variables written in terms of the trap eigenstate basis:

$$z^{(n)}(r, t) = \sum_j z_j^{(n)}(t)\phi_j(r),$$  \hspace{1cm} (10)

$$\rho^{(n)}(r, t) = \sum_{ij} \rho_{ij}^{(n)}(t)\phi_i^*(r)\phi_j(r),$$  \hspace{1cm} (11)

$$\kappa^{(n)}(r, t) = \sum_{ij} \kappa_{ij}^{(n)}(t)\phi_i(r)\phi_j(r).$$  \hspace{1cm} (12)

Real space non-condensate density and non-condensate correlations are, in general, nonlocal functions of two spatial points $\rho(r', r)$ and $\kappa(r', r)$. We only computed these quantities for $r = r'$ in this paper since these are the most physically accessible. Measuring these quantities with $r \neq r'$ involves observing atomic correlations which is much more difficult than photon correlations. In Liouville space notation, the position-dependent $n$th order solution $\tilde{\psi}^{(n)}(r, t)$ can be defined using the relations Eq. (10 - 12) and introducing a $2N(2N + 1) \times 2N(2N + 1)$ square matrix $\tilde{\Upsilon}(r)$:

$$\tilde{\psi}^{(n)}(r, t) \equiv \tilde{\Upsilon}(r)\tilde{\psi}^{(n)}(t)$$  \hspace{1cm} (13)

where

$$\tilde{\Upsilon}(r) = \text{diag} \left[ \tilde{\phi}(r), \tilde{\phi}^*(r), \Phi_\rho(r), \Phi_\kappa(r), \Phi_\rho^*(r), \Phi_\kappa^*(r) \right].$$  \hspace{1cm} (14)

Here “diag[... ]” denotes that $\tilde{\Upsilon}(r)$ is a block diagonal square matrix made of $N \times N$ blocks $\tilde{\phi}(r)$ and $\tilde{\phi}^*(r)$ and $N^2 \times N^2$ blocks $\Phi_\rho(r), \Phi_\kappa(r), \Phi_\rho^*(r), \Phi_\kappa^*(r)$. $\tilde{\phi}(r)$ is a diagonal matrix with the $i$-th diagonal element given by the basis states $\phi_i(r)$, and $\Phi_\rho(r)$ and $\Phi_\kappa(r)$ are also diagonal matrices whose $ij$th diagonal element are given by $\{\Phi_\rho(r)\}_{ij,ij} = \phi_i^*(r)\phi_j(r)$, and $\{\Phi_\kappa(r)\}_{ij,ij} = \phi_i(r)\phi_j(r)$ respectively. The real space variables $z^{(n)}(r, t)$, $\rho^{(n)}(r, t)$, and $\kappa^{(n)}(r, t)$ are finally obtained by summing over the appropriate elements of the vector $\tilde{\psi}^{(n)}(r, t)$:

$$z^{(n)}(r, t) = \sum_{i=1}^n \psi_i^{(n)}(r, t), \quad \rho^{(n)}(r, t) = \sum_{i=2n+1}^{2n+n^2} \psi_i^{(n)}(r, t), \quad \kappa^{(n)}(r, t) = \sum_{i=2n+n^2+1}^{2n+2n^2} \psi_i^{(n)}(r, t).$$  \hspace{1cm} (15)

The position-dependent second order ($n = 2$) response function $K^{(2)}_{\alpha}(t, t_1, t_2, r, r_1, r_2)$ is then defined as follows:

$$\alpha^{(2)}(r, t) = \int K^{(2)}_{\alpha}(t, t_1, t_2, r, r_1, r_2)\, V_j(r_1, t_1)\, V_j(r_2, t_2)\, dt_1\, dt_2\, dr_1\, dr_2,$$  \hspace{1cm} (16)

where $\alpha^{(2)}(r, t)$ are second order solutions, $\alpha = z, \rho, \kappa$. Expression for $K^{(2)}$ is given in Appendix B Eqs. (C1)-(C3). Having found the time domain response $K^{(2)}_{\alpha}(t, t_1, t_2, r_1, r_2)$, the second order susceptibility is obtained by a Fourier transform to the frequency domain:

$$\chi^{(2)}_{\alpha}(\Omega, \Omega_1, \Omega_2, r, r_1, r_2) = \int_0^\infty dt \int_0^\infty dt_1 \int_0^\infty dt_2 K^{(2)}_{\alpha}(t, t_1, t_2, r, r_1, r_2)\, \exp \left( i\Omega t + i\Omega_1 t_1 + i\Omega_2 t_2 \right).$$  \hspace{1cm} (17)

A closed expression for $\chi^{(2)}$ used in our numerical calculations is given in Appendix C Eqs. (D1) - (D3).

### III. TIME DOMAIN RESPONSE

So far, all our results were given in the trap basis, and hold for a general interatomic interaction potential. In the following numerical calculations, we approximate the interatomic potential $V(r - r')$ in Eq. 5 by a contact potential:

$$V(r - r') \rightarrow U_0 \delta(r - r'), \hspace{1cm} U_0 = \frac{4\pi \hbar^2 a}{m},$$  \hspace{1cm} (18)

where $a$ is the $s$-wave scattering length and $m$ is the atomic mass. This is valid because wave functions at ultracold temperatures have very long wavelengths compared to the range of interatomic potential implying that details of the interatomic potential become unimportant. The tetradic matrices $V_{ijkl}$ defined in Eq. 5 are then simply given by:

$$V_{ijkl} = \frac{4\pi \hbar^2 a}{m} \int \phi_i^*(r)\phi_j^*(r)\phi_k(r)\phi_l(r)\, dr.$$  \hspace{1cm} (19)
We consider a 2000 atom one dimensional condensate in a harmonic trap. The parameters used for our numerical calculation of $\vec{\psi}^{(0)}$ are: $U_0 = \frac{4\hbar^2a}{m}$, $0.01$, and temperatures $\hbar\omega_{\text{trap}}/k$ and $10\hbar\omega_{\text{trap}}/k$ where $\omega_{\text{trap}}$ is the trap frequency, $k$ is the Boltzmann constant and the basis set size of $N = 5$ is used. We keep the trap units throughout with 256 grid points for position. The same parameters were used in the calculations of the linear response in Ref. [1].

To solve for the second order response, both the zero\textsuperscript{th} and the first order solutions must be found. Calculation of the zero\textsuperscript{th} order solution from the TIHFB equations is the most numerically involved step, as it requires solving nonlinear coupled equations. Griffin has provided a self-consistent prescription for solving the TIHFB, in terms of the Bogoliubov-de Gennes Equations [6]. We have therefore followed the prescription of Ref. [6] to find the solution to TIHFB. Once the zero\textsuperscript{th} order solution is found, it is straightforward to calculate the first and the second order response functions. The calculation of the eigenvalues of the non-Hermitian matrix $\mathcal{L}^{(2)}$ required for computing the response functions was carried out using the Arnoldi algorithm [31].

In order to provide an indication of the structure of the matrix $\mathcal{L}^{(2)}$, we first plot in Fig. 1 the linear susceptibility $K^{(1)}(\Omega, \mathbf{r} = 0, \mathbf{r}_1 = 0)$ for zero and finite temperatures. Peak positions indicate the resonant frequencies.

An expression for the second order time domain response function is given in Appendix C Eqs. (11) - (18). We have first obtained the numerical solution to TIHFB, $2N(2N + 1) \times 1$ vector $\vec{\psi}^{(0)}$ evaluated at zero and finite temperatures, the $2N(2N + 1) \times 2N(2N + 1)$ matrices $\bar{Y}$, $\bar{U}$, and $\bar{F}$ defined in Eqs. (11), (12), and (14), and the $2N(2N + 1) \times 1$ vector $\bar{\Xi}_K$ defined in Eqs. (11) - (13). Substituting these into Eqs. (14) - (18), the final calculation involves matrix multiplication of $2N(2N + 1) \times 1$ vectors $\vec{\psi}^{(0)}$ and $\bar{\Xi}_K$ with $2N(2N + 1) \times 2N(2N + 1)$ matrices $\bar{Y}$, $\bar{U}$, and $\bar{F}$, and integration over the time variable $t$. $\bar{Y}$ and $\bar{F}$ are constructed in terms of the harmonic oscillator basis states which are calculated numerically from the recursive formula that involves the Gaussian function multiplying the Hermite polynomials [52]. The matrix $\bar{U}$ is calculated using a MATLAB function that uses the Pad\’e approximation for matrix exponentiation [53].

We present the second order response function in the time domain $K^{(2)}(t, t_1, t_2, \mathbf{r}, \mathbf{r}_1, \mathbf{r}_2)$ as a function of $\mathbf{r}$ and $\mathbf{r}_1$ at various times $t, t_1, t_2$ and $t_2$. This provides a way to depict graphically the correlation involving 6 variables $t, t_1, t_2, \mathbf{r}, \mathbf{r}_1,$ and $\mathbf{r}_2$ on a 2 dimensional plot, and gives a “snapshot” of the position-dependent second order correlations across the condensate. The times $t, t_1, t_2$ are respectively the time of detection and the time of the first and second applied short fields, while $\mathbf{r}, \mathbf{r}_1,$ and $\mathbf{r}_2$ denote the corresponding spatial variables. The position-dependence is important since the experimentally produced condensates are mesoscopic in size; in optical spectroscopy, however, the dipole approximation usually applies and consequently the spatial dependence of the response is irrelevant. Fig. 2 shows the absolute value of the second order response functions in the time domain with the time $t_2$ fixed at $t_2 = 0$. Fig. 2 (a) is for the position of perturbation fixed at $\mathbf{r}_2 = 0$ i.e. the center of the trapped atomic cloud while Fig. 2 (b) is for $\mathbf{r}_2 = -5$ at the edge of the cloud. All positions are referred to in harmonic oscillator length units. The plots are for zero temperature condensate at short, intermediate and long times $\{(t, t_1) = (5.89, 2.6), (15.7, 7.2), (31.4, 1.57)\}$ as indicated at the top of each column of figures. The times are given in units of the trap period, $1/\omega_{\text{trap}}$. The top, middle, and bottom rows give the response function for the condensate $z$, non-condensate density $\rho$, and non-condensate correlation $\kappa$ respectively. The dashed circle represents the spatial extent of the trapped BEC. The corresponding plot for a finite temperature BEC at temperature $T = 10\hbar\omega_{\text{trap}}/k$ is displayed in Fig. 3.

The spatially asymmetric functions at $\mathbf{r}_2 = -5$ reverses its shape for $\mathbf{r}_2 = 5$, with similar changes also observed for the $\mathbf{r}_2 = \pm 2.5$ pair. With $\mathbf{r}_2 = \pm 2.5$ which is a point inside the atomic cloud, the contours had markedly less symmetric shape than for $\mathbf{r}_2 = \pm 5$ at the very edge of the atomic cloud. The response functions take a more symmetric shape as time increases. The response functions are preserved at longer times. Comparing Figs. 2 and 3, the response functions show strong temperature dependence with the functions giving distinct contours at different temperatures. The response functions attain spatial symmetry more rapidly at zero temperature owing to the weaker coupling between the variables $z$, $\rho$ and $\kappa$.

IV. FREQUENCY DOMAIN RESPONSE

Using Eq. (11) we have calculated the second order susceptibility. Eq. (12) is also a matrix multiplication involving $2N(2N + 1) \times 1$ vectors $\vec{\psi}^{(0)}$ and $\bar{\Xi}_K(\omega)$ defined in Appendix D Eq. (D10), with the $2N(2N + 1) \times 2N(2N + 1)$ matrices $\bar{Y}$, $\bar{U}(\omega)$, and $\bar{F}$. The Green’s function $\bar{U}(\omega)$ is calculated as follows:

$$\bar{U}(\omega) = \frac{1}{\omega - \mathcal{L}^{(2)} + i\epsilon} = \sum_{\nu} \frac{\xi_{\nu}^* \zeta_{\nu}^*}{\omega - \omega_{\nu} + i\epsilon},$$

(20)

where $\xi_{\nu}$ is the right eigenvector of $\mathcal{L}^{(2)}$ with eigenvalues $\omega_{\nu}$ such that $\mathcal{L}^{(2)} \xi_{\nu} = \omega_{\nu} \xi_{\nu}$ and $\zeta_{\nu}$ are the left eigenvectors of $\mathcal{L}^{(2)}$ such that $\sum_{\nu} \xi_{\nu} \zeta_{\nu}^* = \mathbb{I}$. The eigenvalues $\omega_{\nu}$ of $\mathcal{L}^{(2)}$ were calculated using the Arnoldi algorithm [31].
The absolute value of the second order response function in the frequency domain, \(|K^{(2)}(\Omega_1, \Omega_2, r, r_1, r_2)|\), is displayed in Fig. 4 with the position variable \(r_2\) set at (a) \(r_2 = 0\) at the center of the atomic cloud, and (b) \(r_2 = -5\) at the edge of the atomic cloud. The plots for zero temperature condensate at various frequencies \(\Omega_1\) and \(\Omega_2\) indicated at the top of each column. We chose the frequencies such that \(\Omega_1, \Omega_2, \text{and } \Omega_1 + \Omega_2\) are off-resonant with respect to the eigenvalues of \(\mathcal{L}^{(2)}\) (first column, \(\Omega_1 = 2.23, \Omega_2 = 1.55\)); both \(\Omega_1\) and \(\Omega_2\) are on-resonance while \(\Omega_1 + \Omega_2\) is off-resonant (second column, \(\Omega_1 = 2.2, \Omega_2 = 1.5\)); and finally \(\Omega_1 + \Omega_2\) is on-resonance (\(\Omega_1 = 0.7, \Omega_2 = 1.5\)). As with the previous figures, the top, middle, and bottom rows give the response function for the condensate, non-condensate density, and non-condensate conductivity, respectively. The result for a finite temperature BEC at temperature \(T = 10\hbar\omega_{\text{trap}}/k\) is displayed in Fig. 5. At finite temperature, the resonant frequencies are shifted from the zero temperature counterpart so that the actual frequency combinations used are the following: \(\Omega_1, \Omega_2, \text{and } \Omega_1 + \Omega_2\) off-resonant (\(\Omega_1 = 2.45, \Omega_2 = 1.6\)); both \(\Omega_1\) and \(\Omega_2\) are on-resonance while \(\Omega_1 + \Omega_2\) is off-resonant (\(\Omega_1 = 2.43, \Omega_2 = 1.5\)); and frequencies with \(\Omega_1 + \Omega_2\) are on-resonance (\(\Omega_1 = 0.92, \Omega_2 = 1.5\)).

The susceptibilities become more spatially symmetric as resonant frequencies are matched. The most symmetric function was generated when the sum of two frequencies \(\Omega_1\) and \(\Omega_2\) was on resonance while the least symmetric function resulted when both frequencies were off-resonant. With only one of the frequencies on resonance, a result with an intermediate level of symmetry was observed. The dependence of the response function on \(r_2\) is strongest for the off-resonant case, while the one with \(\Omega_1 + \Omega_2\) on resonance remains more or less unaffected by the changes. As in the time domain response, the asymmetric functions at \(r_2 = -5\) reverses its shape for \(r_2 = 5\). Such change was also observed for the \(r_2 = \pm 2.5\) pair; again, for \(r_2 = \pm 2.5\) which is inside the atomic cloud the contours were found to be less symmetric than the corresponding figures for \(r_2 = \pm 5\).

In Fig. 6 we plot the second order susceptibility at zero temperature as a function of \(\Omega_1\) and \(\Omega_2\) with the position variables set at \(r = r_1 = r_2 = 0\) i.e. at the center of the atomic cloud. The left column shows \(|K^{(2)}(\Omega_1, \Omega_2)|\) i.e. the absolute value of the second order response function. The plot shows only few peaks near the frequency of 1 because the difference between the magnitude of these highest few peaks and the rest of the peaks occurring at other frequencies is too large. This suggests that one should ideally tune into the combination of frequencies \(\Omega_1\) and \(\Omega_2\) corresponding to these highest peaks to observe the maximum second order response experimentally. The second frequency \(\Omega_2\) implies a completely different physics compared to the linear response; \(\Omega_2\) is the frequency of a new harmonic being generated as a result of a strong external perturbation oscillating at frequency \(\Omega_1\). The middle column of Fig. 6 gives \(|K^{(2)}(\Omega_1, \Omega_2)|\) where the largest peaks are scaled down to the magnitude of the smaller peaks present. It clearly shows a number of peaks present at frequencies \(\Omega_1\) and \(\Omega_2\) less than 1, and also shows that with \(\Omega_1\) (\(\Omega_2\)) fixed at 1, there is a pronounced response for many values of \(\Omega_2\) (\(\Omega_1\)). The right column of Fig. 6 shows the logarithm of the left column. This enables the large variations in the magnitude of \(|K^{(2)}(\Omega_1, \Omega_2)|\) to be displayed. The top, middle, and bottom rows give the response function for the condensate, non-condensate density, and non-condensate correlation respectively. Fig. 7 shows the corresponding plot at the finite temperature \(T = 10\hbar\omega_{\text{trap}}/k\). The main difference is that there are more peaks appearing around frequency \(0 < \Omega_i < 1, i = 1, 2\) than at zero temperature.

Since the second order susceptibility \(K^{(2)}(\Omega_1, \Omega_2)\) is by itself a product of several Green’s functions \(U(\omega)\) and linear susceptibilities \(K^{(1)}(\omega), \omega = \Omega_1, \Omega_2, \text{and } \Omega_1 + \Omega_2\), several features of Figs. 6 and 7 are closely related to the linear susceptibility. From Fig. 1, it is clear that the linear susceptibility near frequency of 1 dominates the spectrum at both zero and finite temperatures. It is therefore expected that all second order response \(K^{(2)}(\Omega_1, \Omega_2)\) containing \(\Omega_1 = 1\) or \(\Omega_2 = 1\) component in it will lead a much stronger contribution to the spectrum than those not at \(\Omega_1 = 1\) or \(\Omega_2 = 1\). This explains the features around \(\Omega_1 = 1\) and \(\Omega_2 = 1\). In addition, the linear susceptibility \(K^{(1)}(\omega)\) at finite temperature has more peaks around frequencies \(0 < \omega < 1\) than at zero temperature, as there are more resonances below the frequency of 1 at finite temperature. This leads to the second order susceptibility \(K^{(2)}(\Omega_1, \Omega_2)\) displaying more peaks around frequency \(0 < \Omega_1 < 1\) and \(0 < \Omega_2 < 1\), as illustrated in Fig. 7.

V. NONLINEAR RESPONSE OF FRACTIONAL QUANTUM HALL SYSTEMS

The Fractional Quantum Hall Effect (FQHE) \[20\] for a two-dimensional (2D) electron gas in a strong, perpendicular, external magnetic field is observed through the quantization of the Hall dc conductivity \(\sigma_H(\nu)\) as a function of the filling factor \(\nu \equiv (2\pi n)/(m\omega_c)\), where \(n\) is the mean 2D density of the electrons, \(\omega_c = eB/mc\) is the cyclotron frequency, \(B\) is the magnetic field; \(e\) is the electron charge (we set \(e = h = 1\)). At \(\nu = 1/(2k + 1)\), where \(k\) is an integer \(k = 1, 2, \ldots\), \(\sigma_H(\nu)\) varies in discrete steps, and is given by \(\sigma_H(\nu) = \nu(e^2/2\pi)\).

It has been shown that for \(\nu = 1/(2k + 1)\) the original 2D fermion problem can be mapped into a boson system coupled to a Chern-Simons gauge field added to the time-independent external magnetic field, where the original fermion system and the boson system have the same charge density and Hall conductivity \[21, 22, 23\]. The static Hall conductivity (xy component of the conductivity tensor) calculated for this boson system, \(\sigma_H = \nu(e^2/2\pi)\), coincides with the conductivity obtained by using Laughlin’s ansatz for the many-electron wavefunction \[24\]. For an even
inverse filling number \( \nu = 1/(2k) \) the original fermion problem is mapped onto a composite fermion system coupled to a Chern-Simons gauge field \([22]\). At \( \nu = 1/2 \) this gauge field eliminates the external effective vector potential for the effective composite fermion system \([26,27,28]\). In this section we consider FQHE with the odd inverse denominator of the filling number \( \nu = 1/(2k + 1) \). Using this mapping we can apply our results based on a generalized coherent state (GCS) ansatz for the many-electron wavefunction \([24]\), to compute the second-order response of FQHE. Below we establish the correspondence between the FQHE effective Bose Hamiltonian and the Hamiltonian Eqs. \([1,32,33]\).

We start with the many-electron effective Hamiltonian \( \hat{H} \) where the electron-electron interaction contains Coulomb repulsion \( V(\mathbf{r} - \mathbf{r'}) = 1/|\mathbf{r} - \mathbf{r'}| \), external magnetic vector-potential \( \mathbf{A}(\mathbf{r}) \), and an external electrical scalar potential \( V_f(\mathbf{r},t) \) \([1,33]\):

\[
\hat{H} = \int dt \int d\mathbf{r} \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t) \left( \frac{(-i\nabla_r - e(\mathbf{A}(\mathbf{r}))^2}{2m} - \mu \right) \hat{\psi}_\mathbf{r}(\mathbf{r},t) + \frac{1}{2} \int dt \int d\mathbf{r} \int d\mathbf{r'} \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t) \hat{\psi}_\mathbf{r'}^\dagger (\mathbf{r'},t) V(\mathbf{r} - \mathbf{r'}) \hat{\psi}_\mathbf{r'}(\mathbf{r'},t) \hat{\psi}_\mathbf{r}(\mathbf{r},t) + \int dt \int d\mathbf{r} \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t)V_f(\mathbf{r},t) \hat{\psi}_\mathbf{r}(\mathbf{r},t),
\]

where \( \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t) \) and \( \hat{\psi}_\mathbf{r}(\mathbf{r},t) \) are Fermi creation and annihilation operators; \( m \) is the effective electron band mass and \( \mu \) is the chemical potential.

We next introduce a quasiparticle creation operator \( \hat{\psi}^\dagger (\mathbf{r},t) \) which is related to \( \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t) \) as \([20]\):

\[
\hat{\psi}^\dagger (\mathbf{r},t) = \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t) \exp \left[ -i \nu^{-1} \int d\mathbf{r}' \arg (\mathbf{r} - \mathbf{r'}) \hat{n}(\mathbf{r'},t) \right],
\]

where \( \arg (\mathbf{r} - \mathbf{r'}) \) is the angle between the vector \((\mathbf{r} - \mathbf{r'})\) and the direction of the Hall current (which is perpendicular to both the external magnetic field \( \mathbf{B} = \nabla \times \mathbf{A}(\mathbf{r}) \) and the external electric field \( \mathbf{E} = -\nabla V_f(\mathbf{r},t) \), which are perpendicular to each other); \( \nu^{-1} \) is an odd integer; and \( \hat{n}(\mathbf{r},t) \) is the charge density operator, which is the same for the actual Fermi and artificial Bose system

\[
\hat{n}(\mathbf{r},t) = \hat{\psi}_\mathbf{r}^\dagger (\mathbf{r},t) \hat{\psi}_\mathbf{r}(\mathbf{r},t) = \hat{\psi}^\dagger (\mathbf{r},t) \hat{\psi}(\mathbf{r},t).
\]

The operators \( \hat{\psi}^\dagger (\mathbf{r},t) \) and \( \hat{\psi}(\mathbf{r},t) \) satisfy Bose commutation relations \([21,22]\):

\[
\hat{\psi}(\mathbf{r}) \hat{\psi}^\dagger (\mathbf{r}') - \hat{\psi}^\dagger (\mathbf{r}') \hat{\psi}(\mathbf{r}) = \delta(\mathbf{r} - \mathbf{r'});
\]

\[
\hat{\psi}(\mathbf{r}) \hat{\psi}(\mathbf{r}') - \hat{\psi}(\mathbf{r}') \hat{\psi}(\mathbf{r}) = 0;
\]

\[
\hat{\psi}^\dagger (\mathbf{r}) \hat{\psi}^\dagger (\mathbf{r}') - \hat{\psi}^\dagger (\mathbf{r}') \hat{\psi}^\dagger (\mathbf{r}) = 0.
\]

Using the Bose quasiparticle operators Eq. \([22]\), the Hamiltonian Eq. \([21]\) can be written in a form of the many-boson effective Hamiltonian \( \hat{H} \) by adding the gauge Chern-Simons vector potential \( a(\mathbf{r}) \) \([21,22]\):

\[
\hat{H} = \int dt \int d\mathbf{r} \hat{\psi}^\dagger (\mathbf{r},t) \left( \frac{(-i\nabla_r - e(\mathbf{A}(\mathbf{r}) + a(\mathbf{r}))^2}{2m} - \mu \right) \hat{\psi}(\mathbf{r},t) + \frac{1}{2} \int dt \int d\mathbf{r} \int d\mathbf{r'} \hat{\psi}^\dagger (\mathbf{r},t) \hat{\psi}^\dagger (\mathbf{r'},t) V(\mathbf{r} - \mathbf{r'}) \hat{\psi}(\mathbf{r'},t) \hat{\psi}(\mathbf{r},t) + \int dt \int d\mathbf{r} \hat{\psi}^\dagger (\mathbf{r},t)V_f(\mathbf{r},t) \hat{\psi}(\mathbf{r},t).
\]

In order for the static Hall conductivity of the boson Hamiltonian Eq. \([25]\) in Hartree-Fock-Bogoliubov approximation \([37]\) to coincide with the conductivity obtained by using Laughlin’s ansatz for the many-electron wavefunction \([24]\), the magnetic and gauge potentials in \( k \) space should satisfy \([22]\):

\[
A_\alpha(k) + a_\alpha(k) = \frac{2\pi}{e\nu} \alpha^\beta \frac{i k_\beta}{k^2} (\hat{n}_k - n),
\]

where \( \epsilon^{\alpha\beta} \) is a unit antisymmetric tensor

\[
\epsilon^{\alpha\beta} = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix};
\]
\[ n \] is the mean 2D electron density \[ n = (mn_\text{e})/(2\pi) \], and the Fourier components of the charge density operator \( \hat{n}_k \) with momentum \( k \) is

\[ \hat{n}_k = \sum_k \hat{a}_k \hat{a}_k^\dagger \tag{28} \]

where \( \hat{a}_k \) and \( \hat{a}_k^\dagger \) are Fourier components of \( \hat{\psi}(\mathbf{r}) \) and \( \hat{\psi}(\mathbf{r}) \). Using a basis set of single electron functions \( \phi_i(\mathbf{r}) \), the field operators may be expanded in the form

\[ \hat{\psi}(\mathbf{r}, t) = \sum_i \phi_i(\mathbf{r}) \hat{a}_i(t); \]

\[ \hat{\psi}(\mathbf{r}, t) = \sum_i \phi_i(\mathbf{r}) \hat{a}_i(t), \tag{29} \]

where \( \hat{a}_i^\dagger \) and \( \hat{a}_i \) are Bose operators.

We make the following GCS Hartree-Fock-Bogoliubov ansatz for the time-dependent many-boson wavefunction \[ \Omega \]

\[ |\psi(t)\rangle = \exp \left( \int dt \int d\mathbf{r}(\mathbf{r}, t) \hat{\psi}(\mathbf{r}) + \int dt \int d\mathbf{r} d\mathbf{r}' \beta(\mathbf{r}, \mathbf{r}', t) \hat{\psi}(\mathbf{r}) \hat{\psi}(\mathbf{r}') \right) |\Omega_0\rangle \]

\[ = \exp \left( \sum_i \alpha_i(t) \hat{a}_i^\dagger + \sum_{ij} \beta_{ij}(t) \hat{a}_i \hat{a}_j^\dagger \right)|\Omega_0\rangle, \tag{30} \]

where \( |\Omega_0\rangle \) is an arbitrary normalized reference state with \( \langle \Omega_0 | \Omega_0 \rangle = 1 \).

On the other hand, the operator set of GCS generators of the closed algebra \[ \Omega \] in the exponent of the ansatz Eq. (30) creates an extended Heisenberg-Weyl algebra which may be obtained by a repeated application of the standard boson commutators.

The Hamiltonian that describes the system of many-body interacting bosons is obtained and comes from the basis set expansion of the Hamiltonian Eq. (25) and is given by

\[ \hat{H} = \sum_{ijkl} H_{ijkl} \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_k \hat{a}_l + \sum_{ijkl} V_{ijkl} \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_k \hat{a}_l + \eta \sum_{ij} E_{ij}(t) \hat{a}_i \hat{a}_j, \tag{31} \]

Note that the Hamiltonian Eq. (31) coincides with Eqs. (1) - (5) provided we replace in Eqs. (2) \( H_{ijkl}^{np} \) by \( H_{ijkl} \). In Eq. (31) the single-electron matrix element, \( H_{ijkl} \) given by

\[ H_{ijkl} = \int d\mathbf{r} \phi_i^*(\mathbf{r}) \left( \frac{(-i\nabla_{\mathbf{r}} - e(\mathbf{A}(\mathbf{r}) + \mathbf{a}(\mathbf{r}))}{2m} - \mu \right) \phi_j(\mathbf{r}), \tag{32} \]

\( V_{ijkl} \) is a Coulomb repulsion between two electrons (we put electron charge \( e = 1 \))

\[ V_{ijkl} = \int d\mathbf{r}_1 d\mathbf{r}_2 \phi_i^*(\mathbf{r}_1) \phi_j^*(\mathbf{r}_2) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \phi_k(\mathbf{r}_1) \phi_l(\mathbf{r}_2), \tag{33} \]

and the external electrical field \( E(t) \) field expanded in a basis set is

\[ \sum_{ij} E_{ij}(t) \hat{a}_i \hat{a}_j = \int d\mathbf{r} \phi_i^*(\mathbf{r}) V_j(\mathbf{r}, t) \phi_j(\mathbf{r}) \hat{a}_i^\dagger \hat{a}_j. \tag{34} \]

We can, therefore, apply the results obtained for the Hamiltonian Eqs. (1) - (5) to the FQHE by simply replacing \( H_{ijkl}^{np} \) in Eqs. (2) by \( H_{ijkl} \).

Since in a homogeneous system the momentum is conserved, we can use the plane wave basis, i.e. the eigenfunctions of a momentum \( \mathbf{p} \) \( \phi_p(\mathbf{r}) = S^{-1/2} \exp(-i\mathbf{p}\mathbf{r}) \); where \( S \) is the 2D volume of a system by replacing the indices \( i \) by the momentum \( \mathbf{p} \).

The second-order response is given by Eqs. (D1)-(D3),(D24) in the plane wave basis, provided we use for the Hamiltonian Eqs.(1)-(2) in the Liouvillian Eq. (B6) with \( V_{trap} = 0 \), and replace the inter-particle interaction \( V_{ijkl} \) by

\[ V_{\mathbf{p} - \mathbf{p}'} = \frac{2\pi e^2}{\epsilon|\mathbf{p} - \mathbf{p}'|} + \frac{4\pi^2 n}{m\nu^2|\mathbf{p} - \mathbf{p}'|^2}. \tag{35} \]
where the first term in the r.h.s. represents the 2D Fourier transform component of the Coulomb repulsion and the second term comes from substituting Eq. (26) in Eq. (31). With these substitutions, the FQHE boson Hamiltonian Eq. (31) is reduced to the classical form Eq. (19) and is equivalent to the classical boson Hamiltonian (Eq. (40) in [37]). The double excitations are given by the eigenvalues of the Liouvillian Eq. (B6). The anisotropy in the gauge term Eq. (26) in the Hamiltonian Eq. (31) gives a non-vanishing second-order response, shifting the double-excitation energies with respect to twice the single excitations, as is the case in isotropic systems.

VI. CONCLUSIONS

We have calculated the second order mechanical response functions and susceptibilities of both zero and finite temperature BEC. The systematic, perturbative solution of the TDHFB equations for trapped, atomic BEC enables us to analyze the dynamics of finite temperature BEC. The response of both the non-condensate atoms as well as the condensate were calculated. The calculations apply for a general perturbation of the form $\sum_{ij} \int d^3r \phi_i^*(r)V_f(r,t)\hat{a}_i\hat{a}_j$, where the shape of the external force $V_f(r,t)$ is left arbitrary. The calculated second order response functions were found to show a strong dependence on position and temperature. In the frequency domain, we have observed distinct nonlinear mixing effect, which displays enhanced response at a number of $\Omega_1-\Omega_2$ combinations. The application of the GCS ansatz for the derivation of the second-order response of FQHE 2D electron liquid was presented.

The second order response for BEC has not been reported yet, and most related work is limited to the linear response. As in nonlinear optics, the second and higher order response functions and susceptibilities are clearly expected to be indispensable in characterizing the BEC dynamics in the presence of strong perturbations, and more importantly, in further development of matter-wave nonlinear optics [2].

Acknowledgments

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APPENDIX A: THE TDHFB EQUATIONS

The TDHFB equations of motion that couples $z$, $\rho$, and $\kappa$ are

\begin{align}
  i\hbar \frac{dz}{dt} & = [\mathcal{H}_z + \eta E(t)] z + \mathcal{H}_{zs} z^* , \\
  i\hbar \frac{d\rho}{dt} & = [h, \rho] - (\kappa \Delta^* - \Delta \kappa^*) + \eta [E(t), \rho] , \\
  i\hbar \frac{d\kappa}{dt} & = (h\kappa + \kappa h^*) + (\rho \Delta + \Delta \rho^*) + \Delta + \eta [E(t), \kappa]_+ ,
\end{align}

where $[\ldots]_+$ denotes the anticommutator. Here, $\mathcal{H}_z$, $\mathcal{H}_{zs}$, $h$, and $\Delta$ are $N \times N$ matrices with $n$ being the number of basis wave functions used:

\begin{align}
  [\mathcal{H}_z]_{ij} & = H_{ij} - \mu + \sum_{kl} V_{ijkl} [z_k^* z_l + 2 \rho_{lk}] , \\
  [\mathcal{H}_{zs}]_{ij} & = \sum_{kl} V_{ijkl} \kappa_{kl} , \\
  h_{ij} & = H_{ij} - \mu + 2 \sum_{kl} V_{ijkl} [z_k^* z_l + \rho_{lk}] , \\
  \Delta_{ij} & = \sum_{kl} V_{ijkl} [z_k z_l + \kappa_{kl}] .
\end{align}

$h$ is known as the “Hartree-Fock Hamiltonian” and $\Delta$ as the “pairing field” \cite{2}. $\mu$ is the chemical potential introduced in the Hamiltonian, Eq. (2).
In this Appendix we define the $2N(2N + 1) \times 2N(2N + 1)$ Liouvillian matrix of Eq. (10) As discussed in the main text, it suffices to define the zeroth and the first order matrices $\mathcal{L}^{(0)}$ and $\mathcal{L}^{(1)}$ only, since $\mathcal{L}^{(n)}$ for all $n \geq 1$ is identical. First of all, $\mathcal{L}^{(0)}$ is defined as follows:

$$\mathcal{L}^{(0)} = \begin{pmatrix} H_z^{(0)} - \mu & H_z^{(0)} & 0 & 0 & 0 \\ H_z^{(0)*} & H_z^{(0)*} - \mu & 0 & 0 & 0 \\ 0 & 0 & H^{(-)} & D^\Delta & 0 \\ 0 & 0 & -D^\Delta^* & H^{(+)} & D \\ 0 & 0 & 0 & D^* & H^{(-)*} & D^\Delta^* \\ 0 & 0 & D^* & 0 & -D^\Delta & H^{(+)*} \end{pmatrix},$$  \hspace{1cm} (B1)

where $H_z$ and $H_{z*}$ of $\mathcal{L}^{(0)}$ are as defined in Eqs. (A4, A5) and the remaining $N^2 \times N^2$ submatrices are defined as:

$$H_{ij,mn}^{(-)} = h_{im}^{(0)} \delta_{jn} - h_{mj}^{(0)} \delta_{im}$$  \hspace{1cm} (B2)

$$H_{ij,mn}^{(+)} = h_{im}^{(0)} \delta_{jn} + h_{mj}^{(0)} \delta_{im} + V_{ijmn}$$  \hspace{1cm} (B3)

$$D_{ij,mn} = \Delta_{im}^{(0)} \delta_{jn}$$  \hspace{1cm} (B4)

$$D_{ij,mn}^\Delta = -\Delta_{nj}^{(0)} \delta_{im}$$  \hspace{1cm} (B5)

with $h^{(0)}$ and $\Delta^{(0)}$ being the Hartree-Fock Hamiltonian and the pairing field defined in Eqs. (A6, A7).

For higher orders $n \geq 1$ (in particular, $n = 2$),

$$\mathcal{L}^{(n)} = \mathcal{L}^{(0)} + \mathcal{L'},$$  \hspace{1cm} (B6)

where $\mathcal{L}^{(0)}$ is the zero’th order matrix defined above, and

$$\mathcal{L'} = \begin{pmatrix} \mathcal{V}^{zz_1} & \mathcal{H}^{zz_2} & \mathcal{V}^{z_1} & \mathcal{V}^{z_2} & 0 & 0 \\ \mathcal{V}^{z_2*} & \mathcal{V}^{zz_1*} & 0 & 0 & \mathcal{V}^{z_1*} & \mathcal{V}^{z_2*} \\ \mathcal{V}^{p_1*} & \mathcal{W}^{ph} & \mathcal{W}^{p_2} & \mathcal{W}^{p_1} & 0 & \mathcal{W}^{p_2} \\ \mathcal{V}^{p_2*} & \mathcal{W}^{p_1} & \mathcal{W}^{p_2} & \mathcal{W}^{p_1} & 0 & \mathcal{W}^{p_2} \\ \mathcal{V}^{\nu_1*} & \mathcal{W}^{\nu_1h} & \mathcal{W}^{\nu_1} & \mathcal{W}^{\nu_1} & 0 & \mathcal{W}^{\nu_1h} \\ \mathcal{V}^{\nu_2*} & \mathcal{W}^{\nu_1h} & \mathcal{W}^{\nu_2} & \mathcal{W}^{\nu_1} & 0 & \mathcal{W}^{\nu_2h} \end{pmatrix}. \hspace{1cm} (B7)

The set of $N \times N$ submatrices $\mathcal{V}^{zz_1}$ and $\mathcal{V}^{zz_2}$, $N \times N^2$ submatrices $\mathcal{V}^{z_1}$ and $\mathcal{V}^{z_2}$, $N^2 \times N$ submatrices $\mathcal{V}^{p_1}$, $\mathcal{V}^{p_2}$, $\mathcal{V}^{\nu_1}$, and $\mathcal{V}^{\nu_2}$, and $N^2 \times N^2$ component submatrices $\mathcal{W}^{ph}$, $\mathcal{W}^{p_1h}$, $\mathcal{W}^{p_2h}$, $\mathcal{W}^{p_1}$, $\mathcal{W}^{p_2}$, $\mathcal{W}^{\nu_1h}$, $\mathcal{W}^{\nu_2h}$, and $\mathcal{W}^{\nu_1}$ of $\mathcal{L'}$ are given as follows:

$$\mathcal{V}^{zz_1}_{i,l} = \sum_{kr} V_{iklr} z^{(0)}_k z^{(0)}_r$$  \hspace{1cm} (B8)

$$\mathcal{V}^{z_1}_{i,k} = 2 \sum_r V_{iklr} z^{(0)}_r$$  \hspace{1cm} (B9)

$$\mathcal{V}^{p_1}_{i,j} = \sum_{kr} V_{iklr} z^{(0)*}_k p^{(0)*}_r$$  \hspace{1cm} (B10)

$$\mathcal{V}^{p_2}_{i,j} = \sum_{lr} V_{iklr} z^{(0)*}_l p^{(0)}_r$$  \hspace{1cm} (B11)

$$\mathcal{V}^{\nu_1}_{i,j} = \sum_{lr} V_{iklr} z^{(0)*}_k z^{(0)*}_l$$  \hspace{1cm} (B12)

$$\mathcal{W}^{ph}_{ij,k} = 2 \sum_r V_{iklr} p^{(0)}_r - V_{iklr} p^{(0)*}_r$$  \hspace{1cm} (B13)
\[ W_{ij,kl}^\alpha = \sum_r V_{ikl}^\alpha \kappa_{ij}^{(0)} \quad W^\alpha_{ij,kl} = \sum_r V_{rkl} \kappa^{(0)}_{ir} \]  
\[ W_{ij,kl}^{\alpha \Delta} = \sum_r V_{rkl} \kappa^{(0)*}_{ij} \quad W^{\alpha \Delta}_{ij,kl} = \sum_r V_{rjl} \kappa^{(0)*}_{ir} \]  

(15)
(16)

In addition, the \(2N(2N+1)\times 1\) vector \(\zeta(t)\) is defined as follows:

\[ \zeta(t) = \begin{pmatrix} E(t) & 0 & 0 & 0 & 0 & 0 \\ 0 & E^*(t) & 0 & 0 & 0 & 0 \\ 0 & 0 & e^-(t) & 0 & 0 & 0 \\ 0 & 0 & 0 & e^+(t) & 0 & 0 \\ 0 & 0 & 0 & 0 & [e^-(t)]^* & 0 \\ 0 & 0 & 0 & 0 & 0 & [e^+(t)]^* \end{pmatrix}, \]  

(17)

where \(E(t)\) is as defined in Eq. (4), and we have further defined the \(N^2 \times N^2\) submatrices

\[ e^{(\pm)}_{ij,kl} = E_{ik}(t)\delta_{jl} \pm E_{ij}(t)\delta_{ik}. \]  

(18)

**APPENDIX C: SECOND ORDER RESPONSE FUNCTIONS**

In this Appendix, we first summaries the final result for the response functions, and then provide a more detailed derivation. In the last subsection of this Appendix, we also show an alternative form for the response functions written in the basis of the eigenvectors of the Liouvillian.

1. **Final expression**

The second order response functions for the condensate, the non-condensate density and correlation that we use in our numerical calculations are given by Eqs. (C1) - (C3) as follows:

\[ K^{(2)}_{z}(t_1, t_2, r_1, r_2) = \sum_{i=1}^{N} \tilde{\Upsilon}(r) \left( \tilde{K}^{(2)}_{I}(t_1, t_2, r_1, r_2) + \tilde{K}^{(2)}_{II}(t_1, t_2, r_1, r_2) \right) \]  

(11)

\[ K^{(2)}_{\rho}(t_1, t_2, r_1, r_2) = \sum_{i=2N+1}^{2N+N^2} \tilde{\Upsilon}(r) \left( \tilde{K}^{(2)}_{I}(t_1, t_2, r_1, r_2) + \tilde{K}^{(2)}_{II}(t_1, t_2, r_1, r_2) \right) \]  

(12)

\[ K^{(2)}_{\kappa}(t_1, t_2, r_1, r_2) = \sum_{i=2N+1}^{2N+N^2} \tilde{\Upsilon}(r) \left( \tilde{K}^{(2)}_{I}(t_1, t_2, r_1, r_2) + \tilde{K}^{(2)}_{II}(t_1, t_2, r_1, r_2) \right) \]  

(13)

where \(2N(2N+1)\times 2N(2N+1)\) matrix \(\tilde{\Upsilon}(r)\) was defined in Eq. (14), and the \(2N(2N+1)\times 1\) vectors \(\tilde{K}^{(2)}_{I}\) and \(\tilde{K}^{(2)}_{II}\) are defined as follows:

\[ \tilde{K}^{(2)}_{I}(t_1, t_2, r_1, r_2) = \mathcal{U}(t - t_1) \tilde{\Phi}(r_1) \mathcal{U}(t_1 - t_2) \tilde{\Phi}(r_2) \tilde{\psi}(0), \]  

(14)

\[ \tilde{K}^{(2)}_{II}(t_1, t_2, r_1, r_2) = \int_0^t d\tau \mathcal{U}(t - \tau) \tilde{\Xi}(\tau - t_1, \tau - t_2, r_1, r_2), \]  

(15)

where

\[ \mathcal{U}(t - t') = \exp \left[ -\frac{i}{\hbar} \mathcal{L}^{(2)}(t - t') \right], \]  

(16)
\[
\tilde{\Phi}(\mathbf{r}') = \text{diag}\left[\Phi(\mathbf{r}'), \Phi^*(\mathbf{r}'), \Phi^{(-)}(\mathbf{r}'), \Phi^{(+)}(\mathbf{r}'), \Phi^{(-)*}(\mathbf{r}'), \Phi^{(+)*}(\mathbf{r}')\right].
\]

\(\tilde{\Phi}(\mathbf{r}')\) is therefore a \(2N(2N + 1) \times 2N(2N + 1)\) block diagonal square matrix with the blocks consisting of \(N \times N\) square matrices

\[
[\Phi(\mathbf{r})]_{ij} = \phi_i^*(\mathbf{r})\phi_j(\mathbf{r})
\]

and \(N^2 \times N^2\) square matrices

\[
[\Phi(\mathbf{r})]_{ij, mn} = \phi_i^*(\mathbf{r})\phi_m(\mathbf{r})\delta_{jn} \pm \phi_n^*(\mathbf{r})\phi_j(\mathbf{r})\delta_{im}.
\]

The vector \(\Xi_K\) of Eq. (C3) may be written

\[
\Xi_K = [Z_K, Z_K^*, R_K, K_K, R_K^*, K_K^*]^T
\]

with the \(N \times 1\) matrix \(Z_K\) and \(N^2 \times 1\) matrices \(R_K, K_K\) given as follows:

\[
\begin{aligned}
[Z_K]_{ij} &= \sum_{klr} V_{iklr} \left( [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} z_{i}^{(0)} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{r} 
+ z_{k}^{(0)} [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{lj} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{r} 
+ 2 [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{r} 
+ 2 [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{jl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{r} \right) 
\end{aligned}
\]

\[
[\mathcal{R}_K]_{ij} = \sum_{rlk} V_{iklr} [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{rl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ \sum_{rlk} V_{irkl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ 2 [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ 2 [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} \]

\[
[K_K]_{ij} = \sum_{rlk} V_{iklr} [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{rl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ \sum_{rlk} V_{irkl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ 2 [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ 2 [\tilde{K}_z^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_z^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} \]

\[
[\mathcal{K}_K]_{ij} = \sum_{rlk} V_{iklr} [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{rl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ \sum_{rlk} V_{irkl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ 2 [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} 
+ 2 [\tilde{K}_p^{(1)}(\mathbf{r} - t_1, \mathbf{r}_1)]_{kl} [\tilde{K}_p^{(1)}(\mathbf{r} - t_2, \mathbf{r}_2)]_{lj} \]

\]
where the vectors \( \vec{K}_z^{(1)}(\tau-t_1, \mathbf{r}_1), \vec{K}_\rho^{(1)}(\tau-t_1, \mathbf{r}_1) \) and \( \vec{K}_\kappa^{(1)}(\tau-t_1, \mathbf{r}_1) \) found in Eqs. (C12) are defined as

\[
\vec{K}_\alpha^{(1)}(\tau-t_1, \mathbf{r}_1) = \mathcal{U}^{(\alpha)}(\tau-t_1)\vec{\Phi}(t_1)\psi^{(0)}(t), \quad \alpha = z, \rho, \kappa, \quad i = 1, 2.
\]

\( \mathcal{U}^{(\alpha)}(\tau-t_1) \) are the submatrices of \( \mathcal{U}(t-t_1) \) in Eqs. (C11) which, in turn, are stacked on top of submatrices \( \mathcal{U}^{(z)}(\tau-t_1) \) which, in turn, are stacked on top of submatrices \( \mathcal{U}^{(\gamma)}(\tau-t_1) \).

where to be noted that \( \vec{K}_z^{(1)}(\tau-t_1, \mathbf{r}_1) \) as defined here are \( N \times 1 \) and \( N^2 \times 1 \) vectors, not scalar quantities obtained by integrating the scalar function \( K^{(1)}_\alpha(\tau-t_1, \mathbf{r}, \mathbf{r}_1) \) over \( \mathbf{r} \).

### 2. Derivation

Writing the second order solution to TDHFB explicitly, we have

\[
\psi^{(2)}(t) = \frac{1}{\sqrt{i\hbar}} \int_0^t \exp \left[ -i \frac{\hbar}{\hbar} \mathcal{L}^{(2)}(t-t_1) \right] \vec{\Gamma}(t_1) dt_1
\]

\[
\equiv \frac{1}{\sqrt{i\hbar}} \int_0^t \mathcal{U}(t-t_1) \vec{\Gamma}(t_1) dt_1
\]

\[
\vec{\Gamma}(t_1) = \zeta(t_1)\psi^{(1)}(t_1) + \vec{\Xi}(t_1).
\]

i.e. for the second order response, \( \chi^{(2)}(t) \equiv \zeta(t)\psi^{(1)}(t) + \vec{\Xi}(t) \) in Eq. (D) where \( \zeta(t) \) is given in Appendix B and \( \vec{\Xi}(t) \) is a \( 2N(2N+1) \times 1 \) vector originating from the terms in the expansion which are made up of products of two first order variables i.e. \( z^{(1)}, \rho^{(1)} \) and \( \kappa^{(1)} \). The vector \( \vec{\Xi}(t) \) can be written as \( \vec{\Xi}(t) = [\mathcal{Z}, \mathcal{Z}\mathcal{R}, \mathcal{K}, \mathcal{K}\mathcal{R}, \mathcal{K}^*]^T \) with the \( N \times 1 \) matrix \( \mathcal{Z} \), and \( N^2 \times 1 \) matrices \( \mathcal{R}, \mathcal{K} \) given as follows:

\[
\mathcal{Z}_i = \sum_{klr} V_{ikl} \left[ z^{(1)}_k z^{(0)}_l z^{(1)}_r + \rho^{(1)}_k \rho^{(0)}_l z^{(1)}_r + \rho^{(1)}_k \rho^{(0)}_l z^{(1)}_r + 2\rho^{(00)}_k \rho^{(1)}_l z^{(1)}_r + \kappa^{(1)}_k \kappa^{(1)}_l \right],
\]

\[
\mathcal{R}_{ij} = 2 \sum_{krl} V_{ikl} \rho^{(1)}_j \rho^{(1)}_l - V_{ikl} \rho^{(0)}_j \rho^{(1)}_l + \sum_{rkl} V_{ijk} \kappa^{(1)}_l \kappa^{(1)}_l + V_{irk} \kappa^{(1)}_l \kappa^{(1)}_l + 2 \sum_{rkl} V_{ikl} z^{(1)}_k z^{(0)}_l \rho^{(1)}_l - V_{ikl} z^{(1)}_k z^{(0)}_l \rho^{(1)}_l + 2 \sum_{rkl} V_{ikl} z^{(1)}_k z^{(0)}_l \rho^{(1)}_l - V_{irkl} z^{(1)}_k z^{(0)}_l \rho^{(1)}_l + \sum_{rkl} V_{irk} \kappa^{(1)}_l \kappa^{(1)}_l - V_{irk} \kappa^{(1)}_l \kappa^{(1)}_l.
\]
\[
\begin{align*}
\mathcal{K}_{ij} &= 2 \sum_{rkl} V_{ikl} \kappa_{ir} \rho_k \left(1\right) + V_{rlkj} \kappa_{kl} \kappa_{ij} \left(1\right) + \sum_{rkl} V_{ikl} \kappa_{kl} \rho_{ir} \left(1\right) + V_{rklj} \kappa_{kl} \rho_{ir} \\
&+ 2 \sum_{rkl} V_{ikl} \left[ z_k \kappa_{ir} \zeta_i \left(1\right) + z_k \zeta_i \kappa_{ir} \left(1\right) \right] + V_{rlkj} \zeta_i \kappa_{ir} \left(1\right) + \kappa_{ij} \left(1\right) \kappa_{ir} \left(0\right) \\
&+ \sum_{rkl} V_{rklj} \left[ z_k \kappa_{ij} \zeta_l \left(1\right) + z_k \zeta_l \kappa_{ij} \left(1\right) \right] \rho_{ir} \left(1\right) + V_{rklj} \zeta_l \kappa_{ij} \left(1\right) \rho_{ir} \left(1\right) \\
&+ \sum_{rkl} V_{rklj} \kappa_{ij} \left(0\right) + V_{rklj} \kappa_{ij} \left(1\right) \rho_{ir} \left(0\right) + \sum_{kl} V_{rklj} \kappa_{ij} \left(1\right) \zeta_i \left(1\right).
\end{align*}
\]

Casting Eq. \((C17)\) in the form
\[
\tilde{\psi}^{(2)}(r, t) = \int \tilde{K}^{(2)}(t, t_1, t_2, r, r_1 r_2) V_f(r_1, t_1) V_f(r_2, t_2) d^3 r_1 dt_1 d^3 r_2 dt_2
\]

involves rewriting \(\bar{I}(t)\) of Eq. \((C18)\) in the position dependent form:
\[
\bar{I}(t_1) = \frac{1}{\sqrt{\hbar}} \int_0^{t_1} dt_2 \int dr_1 dr_2 \left[ \Phi(r_1) \tilde{K}^{(1)}(t_1, t_2, r_2) \right] V_f(r_1, t_1) V_f(r_2, t_2) + \int_0^{t_1} dt_3 \int dr_2 dr_3 \left[ \tilde{\Xi}_K(t_1 - t_2, t_1 - t_3; r_2, r_3) \right] V_f(r_2, t_2) V_f(r_3, t_3),
\]

where \(\tilde{K}^{(1)}(t_1, t_2, r_2)\) is the linear response function for the combined variables \(z, \rho\) and \(\kappa\):
\[
\tilde{K}^{(1)}(t_1, t_2, r_2) = \mathcal{U}(t_1 - t_2) \tilde{\Phi}(r_2) \Phi^{(0)},
\]

and the \(2N(2N + 1) \times 1\) column vector \(\tilde{\Xi}_K(t_1 - t_2, t_1 - t_3; r_2, r_3)\) has already been defined above in Eqs. \((C10)\) \((C13)\). \(\tilde{\Xi}_K\) is derived from the vector \(\tilde{\psi}\) such that the components of the linear response vector \(\tilde{K}^{(1)}(t_1, t_2, r_2)\) i.e. \(K^{(1)}_z(t_1, t_2, r_2)\), \(K^{(1)}_{\rho}(t_1, t_2, r_2)\) and \(K^{(1)}_{\kappa}(t_1, t_2, r_2)\) defined in Eq. \((C14)\) replace, respectively, \(z^{(1)}(t_1)\), \(\rho^{(1)}(t_1)\), and \(\kappa^{(1)}(t_1)\) in \(\tilde{\Xi}(t)\).

Using these results, the position-dependent time domain second-order response function for the combined variables \(z, \rho, \) and \(\kappa\) may finally be written as
\[
\tilde{K}^{(2)}(t, t_1, t_2, r, r_1, r_2) = \tilde{\Theta}(r) \left[ \tilde{K}^{(2)}_z + \tilde{K}^{(2)}_f \right],
\]

where, after the change of variables \(t_1 \to \tau, \ t_2 \to t_1, \ t_3 \to t_2, \ K^{(2)}_f\) and \(K^{(2)}_f\) are given by
\[
\begin{align*}
\tilde{K}^{(2)}_f(t, t_1, t_2, r_1, r_2) &= \mathcal{U}(t - t_1) \tilde{\Phi}(r_1) \mathcal{U}(t_1 - t_2) \tilde{\Phi}(r_2) \Phi^{(0)}, \\
\tilde{K}^{(2)}_f(t, t_1, t_2, r_1, r_2) &= \int_0^t d\tau \mathcal{U}(t - \tau) \tilde{\Xi}_K(\tau - t_1, \tau - t_2, r_1, r_2).
\end{align*}
\]

The position-dependent second order response functions for the condensate, the non-condensate density and correlation are given by summing over appropriate indices of the vector \(\tilde{K}^{(2)}\), Eq. \((C20)\):
\[
\begin{align*}
K^{(2)}_z(t, t_1, t_2, r, r_1, r_2) &= \sum_{i=1}^{\sqrt{2}N} \tilde{\Theta}(r) \left[ \tilde{K}^{(2)}_f(t, t_1, t_2, r_1, r_2) + \tilde{K}^{(2)}_f(t, t_1, t_2, r_1, r_2) \right], \\
K^{(2)}_f(t, t_1, t_2, r, r_1, r_2) &= \sum_{i=2N+1}^{2N+N^2} \tilde{\Theta}(r) \left[ \tilde{K}^{(2)}_f(t, t_1, t_2, r_1, r_2) + \tilde{K}^{(2)}_f(t, t_1, t_2, r_1, r_2) \right].
\end{align*}
\]
\begin{equation}
K^{(2)}_{\kappa}(t,t_1,t_2,r_1,r_2) = \sum_{i=2N+2N^2+1}^{2N+2N^2} \left[ \tilde{\varPsi}(r) \left( \tilde{K}^{(2)}_{I}(t,t_1,t_2,r_1,r_2) + \tilde{K}^{(2)}_{II}(t,t_1,t_2,r_1,r_2) \right) \right]_i \tag{C31}
\end{equation}

where $\tilde{K}^{(2)}_{I}$ and $\tilde{K}^{(2)}_{II}$ are defined in Eqs. (C6) - (C8), and $\tilde{\varPsi}, \hat{U}, \tilde{\varPhi}$ and $\bar{\Gamma}_K$ are defined in Eqs. (H1), (H4), (H7), and (H10 - H12) respectively.

3. Alternative form for the response function

To discuss the response functions in the frequency domain and to understand the physical processes involved, it is useful to expand the response functions in the basis of the eigenvectors $\xi_{\nu}$, of matrix $L^{(2)}$ such that $L^{(2)} \xi_{\nu} = \omega_{\nu} \xi_{\nu}$, $\nu = 1,2,\ldots,2N(2N+1)$. We define the Green's function

\begin{equation}
G_{\nu}(t-t') = \exp \left[ -\frac{i}{\hbar} \omega_{\nu}(t-t') \right], \tag{C32}
\end{equation}

and the expansion coefficients $\mu_{\nu}, \eta_{\nu}(r)$, and $\delta_{\nu}(r)$ such that

\begin{equation}
\tilde{\varPsi}^{(0)} = \sum_{\nu=1}^{2N(2N+1)} \mu_{\nu} \tilde{\xi}_{\nu}, \quad \hat{\varPhi}(r) \tilde{\xi}_{\nu} = \sum_{\nu=1}^{2N(2N+1)} \eta_{\nu}(r) \tilde{\xi}_{\nu}, \quad \text{and} \quad \tilde{\varPsi}(r) \tilde{\xi}_{\nu} = \sum_{\nu=1}^{2N(2N+1)} \delta_{\nu}(r) \tilde{\xi}_{\nu}. \tag{C33}
\end{equation}

In the basis of these eigenstates, $\tilde{\xi}_{\nu}$, the result is Eqs. (C20) - (C31) but with

\begin{equation}
K^{(2)}_{I}(t,t_1,t_2,r_1,r_2) = \sum_{\nu=1}^{2N(2N+1)} K^{(2)}_{I,\nu}(t,t_1,t_2,r_1,r_2) \tilde{\xi}_{\nu}, \tag{C34}
\end{equation}

\begin{equation}
K^{(2)}_{II}(t,t_1,t_2,r_1,r_2) = \sum_{\nu=1}^{2N(2N+1)} K^{(2)}_{II,\nu}(t,t_1,t_2,r_1,r_2) \tilde{\xi}_{\nu}, \tag{C35}
\end{equation}

where

\begin{equation}
K^{(2)}_{I,\nu}(t,t_1,t_2,r_1,r_2) = \sum_{\nu'=1}^{2N(2N+1)} \eta_{\nu}(r_1) \eta_{\nu'}(r_2) G_{\nu}(t-t_1) \mu_{\nu'} G_{\nu'}(t_1-t_2), \tag{C36}
\end{equation}

and

\begin{equation}
K^{(2)}_{II,\nu}(t,t_1,t_2,r_1,r_2) = \sum_{\nu',\nu''=1}^{2N(2N+1)} \int_0^t d\tau G_{\nu}(t-\tau) F \left[ \eta_{\nu}(r_1) \eta_{\nu'}(r_2) \mu_{\nu'} \mu_{\nu''} G_{\nu'}(\tau-t_1) G_{\nu''}(\tau-t_2) \right], \tag{C37}
\end{equation}

where $F$ is the function given according to the expression for $\bar{\Gamma}_K$ but written in terms of $\mu_{\nu}, \eta_{\nu},$ and $G_{\nu}$ with $G_{\nu}(t-t')$, $\mu_{\nu}$, and $\eta_{\nu}$ as defined in Eqs. (C32 - C33).

**APPENDIX D: SECOND ORDER SUSCEPTIBILITIES**

1. Final expression

The final result that we use for our numerical calculation for the condensate, non-condensate density and the non-condensate correlations are:

\begin{equation}
K^{(2)}_{\kappa}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{i=1}^{N} \left[ \tilde{\varPhi}(r) \left( \tilde{K}^{(2)}_{I}(\Omega_1, \Omega_2, r, r_1, r_2) + \tilde{K}^{(2)}_{II}(\Omega_1, \Omega_2, r, r_1, r_2) \right) \right]_i \tag{D1}
\end{equation}

\begin{equation}
K^{(2)}_{\rho}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{i=2N+1}^{2N+N^2} \left[ \tilde{\varPhi}(r) \left( \tilde{K}^{(2)}_{I}(\Omega_1, \Omega_2, r, r_1, r_2) + \tilde{K}^{(2)}_{II}(\Omega_1, \Omega_2, r, r_1, r_2) \right) \right]_i \tag{D2}
\end{equation}
\[ K^{(2)}_{\kappa}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r_1, r_2) = \sum_{i=2N+1}^{2N+2N^2} \left[ \tilde{\Upsilon}(r) \left( \tilde{K}^{(2)}_j(\Omega_1, \Omega_2, r_1, r_2) + \tilde{K}^{(2)}_{ij}(\Omega_1, \Omega_2, r_1, r_2) \right) \right]_i. \]  
(D3)

where

\[ \tilde{K}^{(2)}_j(\Omega_1, \Omega_2, r_1, r_2) = -\frac{1}{4\pi^2} \tilde{\Upsilon}(r) U(\Omega_1 + \Omega_2) \tilde{\Phi}(r_1) U(\Omega_2) \tilde{\Phi}(r_2) \psi^{(0)}, \]  
(D4)

and

\[ \tilde{K}^{(2)}_{ij}(\Omega_1, \Omega_2, r_1, r_2) = -\frac{1}{8\pi^2} \tilde{\Upsilon}(r) U(\Omega_1 + \Omega_2) \tilde{\xi}_{\kappa}(\Omega_1, \Omega_2, r_1, r_2). \]  
(D5)

Here, \( \tilde{\Upsilon}(r) \) and \( \tilde{\Phi}(r) \) are as defined in Eqs. \( \text{(4)} \) and \( \text{(7)} \) and

\[ U(\omega) \equiv \frac{1}{\omega - L^{(2)} + i\epsilon}. \]  
(D6)

In addition, the vector \( \tilde{\xi}_{\kappa}(\Omega_1, \Omega_2, r_1, r_2) \) of Eq. \( \text{(D5)} \) is may be written as

\[ \tilde{\xi}_{\kappa}(\Omega_1, \Omega_2, r_1, r_2) = [\tilde{Z}_{\kappa}, \tilde{Z}^{*}_{\kappa}, \tilde{R}_{\kappa}, \tilde{K}_{\kappa}, \tilde{K}^{*}_{\kappa}, \tilde{K}^{(2)}_{\kappa}]^T \]  
(D7)

with the \( N \times 1 \) matrix \( \tilde{Z}_{\kappa} \) and \( N^2 \times 1 \) matrices \( \tilde{R}_{\kappa}, \tilde{K}_{\kappa} \) given as follows:

\[
\left[ \tilde{Z}_{\kappa} \right]_i = \sum_{k,l,r} V_{klr} \left[ \left[ \tilde{K}^{(1)}_z(\Omega_1, r_1) \right]_k [\tilde{Z}^{(0)}_{\kappa}(\Omega_2, r_2)]_l + \left[ \tilde{K}^{(1)}_z(\Omega_1, r_1) \right]_k [\tilde{Z}^{(0)}_{\kappa}(\Omega_2, r_2)]_l \right] z^{(0)}_r
+ \left[ z^{(0)}_r \right] \left[ \tilde{K}^{(1)}_z(\Omega_1, r_1) \right]_k \left[ \tilde{Z}^{(1)}_{\kappa}(\Omega_2, r_2) \right]_l + \left[ \tilde{Z}^{(1)}_{\kappa}(\Omega_1, r_1) \right]_k \left[ \tilde{Z}^{(1)}_{\kappa}(\Omega_2, r_2) \right]_l
+ \left[ \tilde{Z}^{(1)}_{\kappa}(\Omega_1, r_1) \right]_k \left[ \tilde{Z}^{(1)}_{\kappa}(\Omega_2, r_2) \right]_l
\]  
(D8)

\[
\left[ \tilde{R}_{\kappa} \right]_{ij} = 2 \sum_{k,l,r} V_{klr} \left[ \left[ \tilde{K}^{(1)}_{\kappa}^{*}(\Omega_1, r_1) \right]_k \left[ \tilde{K}^{(1)}_z(\Omega_2, r_2) \right]_l + \left[ \tilde{K}^{(1)}_{\kappa}^{*}(\Omega_1, r_1) \right]_k \left[ \tilde{K}^{(1)}_z(\Omega_2, r_2) \right]_l \right] z^{(0)}_r
+ \sum_{k,l,r} V_{klr} \left[ \left[ \tilde{K}^{(1)}_{\kappa}^{*}(\Omega_1, r_1) \right]_k \left[ \tilde{K}^{(1)}_z(\Omega_2, r_2) \right]_l \right] [\tilde{K}^{(0)}_{\kappa}(\Omega_2, r_2)]_l + \sum_{k,l,r} V_{klr} \left[ \left[ \tilde{K}^{(1)}_{\kappa}^{*}(\Omega_1, r_1) \right]_k \left[ \tilde{K}^{(1)}_z(\Omega_2, r_2) \right]_l \right] [\tilde{K}^{(0)}_{\kappa}(\Omega_2, r_2)]_l
+ \sum_{k,l,r} V_{klr} \left[ \left[ \tilde{K}^{(1)}_{\kappa}^{*}(\Omega_1, r_1) \right]_k \left[ \tilde{K}^{(1)}_z(\Omega_2, r_2) \right]_l \right] \left[ \tilde{K}^{(1)}_{\kappa}(\Omega_2, r_2) \right]_l
\]  
(D9)
\[
\begin{align*}
+ \sum_{ijkl} V_{ijkl} [\tilde{K}_{z}^{(1)}(\Omega_1, r_1) |_{k} z^{(0)} + z^{(0)} | k \tilde{K}_{z}^{(1)}(\Omega_1, r_1)] [\tilde{K}_{\rho}^{(1)}(\Omega_2, r_2)]_{ir} \\
+ V_{ikl} [\tilde{K}_{z}^{(1)}(\Omega_1, r_1) |_{k} z^{(0)} + z^{(0)} | k \tilde{K}_{z}^{(1)}(\Omega_1, r_1)] [\tilde{K}_{\rho}^{(1)}(\Omega_2, r_2)]_{rj} \\
+ \sum_{ijkl} V_{ijkl} [\tilde{K}_{z}^{(1)}(\Omega_1, r_1) k [\tilde{K}_{z}^{(1)}(\Omega_2, r_2)]_{ir} + V_{ikl} [\tilde{K}_{z}^{(1)}(\Omega_1, r_1) k [\tilde{K}_{z}^{(1)}(\Omega_2, r_2)]_{rj} \\
+ \sum_{kl} V_{ijkl} [\tilde{K}_{z}^{(1)}(\Omega_1, r_1) k [\tilde{K}_{z}^{(1)}(\Omega_2, r_2)]_{ir}] (D10)
\end{align*}
\]

0 Similar to Eq. [C14] above, the quantities \( \tilde{K}_{z}^{(1)}(\Omega_1, r_1) \), \( \tilde{K}_{\rho}^{(1)}(\Omega_1, r_1) \) and \( \tilde{K}_{\kappa}^{(1)}(\Omega_1, r_1) \) used in Eqs. [D8, D10] are defined as

\[
\tilde{K}_{\alpha}^{(1)}(\Omega_1, r_1) = U^{(\alpha)}(\Omega_1) \tilde{\Phi}(r_1) \psi^{(0)},
\]

where \( \alpha = z, \rho, \kappa \), and \( i = 1, 2 \). \( U^{(\alpha)}(\Omega_1) \) are the submatrices of \( U(\Omega_1) \) defined in Eq. [D6] such that

\[
U(\Omega_1) = \left[ U^{(z)}(\Omega_1), U^{(z)+}(\Omega_1), U^{(\rho)}(\Omega_1), U^{(\kappa)}(\Omega_1), U^{(\rho)+}(\Omega_1), U^{(\kappa)+}(\Omega_1) \right]^T
\]

where \( U^{(z)}(\Omega_1) \) is an \( N \times 2N(2N + 1) \) submatrix while \( U^{(\gamma)}(\Omega_1) \), \( \gamma = \rho, \kappa, \rho^*, \kappa^* \) is an \( N^2 \times 2N(2N + 1) \) submatrix such that the submatrix \( U^{(z)}(\Omega_1) \) is stacked on top of submatrix \( U^{(\gamma)}(\Omega_1) \) which, in turn, is stacked on top of submatrices \( U^{(\gamma)}(\Omega_1) \). It is to be noted that, as with the time domain example discussed above, \( \tilde{K}_{\alpha}^{(1)}(\Omega_1, r_1) \) as defined here are \( N \times 1 \) and \( N^2 \times 1 \) vectors, not scalar quantities obtained by integrating the scalar function \( \tilde{K}_{\alpha}^{(1)}(\Omega_1, r, r_1) \) over \( r \).

2. Derivation

The second order response function in frequency is given by the Fourier Transform of the time domain counterpart:

\[
\tilde{K}_{ij}^{(2)}(\Omega, \Omega_1, \Omega_2, r, r_1, r_2) = \int_0^\infty dt dt_1 \tilde{\Upsilon}(r) \left[ \tilde{K}_{ij}^{(2)} + \tilde{K}_{jj}^{(2)} \right] \exp(i \Omega t + i \Omega_1 t_1 + i \Omega_2 t_2) \]

\[
= \tilde{\Upsilon}(r) \left[ \tilde{K}_{ij}^{(2)}(\Omega, \Omega_1, \Omega_2, r_1, r_2) + \tilde{K}_{jj}^{(2)}(\Omega, \Omega_1, \Omega_2, r_1, r_2) \right], \quad (D14)
\]

where \( \tilde{K}_{ij}^{(2)}(\Omega, \Omega_1, \Omega_2, r_1, r_2) \) and \( \tilde{K}_{jj}^{(2)}(\Omega, \Omega_1, \Omega_2, r_1, r_2) \) are the Fourier transforms of the time domain expressions Eq. [C24]- [C25].

Using the fact that the matrices \( U(t-t_1) \) are the Green’s functions with an implicit Heaviside step function in time i.e. \( U(t-t_1) \equiv \theta(t-t_1) U(t-t_1) \) such that

\[
\theta(t) U(t) = \frac{1}{2 \pi i} \int_{-\infty}^{\infty} \frac{1}{\omega - C^{(2)} + i \epsilon} \exp(-i \omega t)
\]

\[
= \int_{-\infty}^{\infty} d\omega U(\omega) \exp(-i \omega t), \quad (D16)
\]

we have

\[
\tilde{K}_{ij}^{(2)}(\Omega, \Omega_1, \Omega_2, r, r_1, r_2) = -\frac{1}{4 \pi^2} \int_{-\infty}^{\infty} d\omega d\omega' \tilde{\Upsilon}(r) U(\omega) \tilde{\Phi}(r_1) U(\omega') \tilde{\Phi}(r_2) \psi^{(0)} \delta(\Omega - \omega)
\]

\[
\times \delta(\Omega_1 + \omega - \omega') \delta(\Omega_2 + \omega'). \quad (D17)
\]

This implies that \( \omega' = -\Omega_2, \omega = -\Omega_1 - \Omega_2, \text{ and } \Omega = -\Omega_1 - \Omega_2; \)

\[
\tilde{K}_{ij}^{(2)}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = -\frac{1}{4 \pi^2} \tilde{\Upsilon}(r) U(\Omega_1 + \Omega_2) \tilde{\Phi}(r_1) U(\Omega_2) \tilde{\Phi}(r_2) \psi^{(0)}.
\]

In addition we have,

\[
\tilde{K}_{ij}^{(2)}(\Omega, \Omega_1, \Omega_2, r, r_1, r_2) = -\frac{1}{8 \pi^3 i} \int_{-\infty}^{\infty} d\omega d\omega' d\omega'' \tilde{\Upsilon}(r) U(\omega) \tilde{\Xi}(\omega, \omega', r_1, r_2)
\]

\[
\times \delta(\omega - \omega' - \omega'') \delta(\Omega - \omega) \delta(\Omega_1 + \omega') \delta(\Omega_2 + \omega''). \quad (D19)
\]
We are able to write the Fourier Transform for $\tilde{\Xi}_K(t)$ in Eq. (D19) since the function $\tilde{\Xi}_K(t)$ is made up of terms which are simply products of two Green’s functions at different times. Eq. (D19) implies $\omega' = -\Omega_1$, $\omega'' = -\Omega_2$, $\omega = \omega' + \omega'' = -\Omega_1 - \Omega_2$, $\Omega = \omega$ so that

$$K^{(2)}_{II}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = -\frac{1}{8\pi^2} \tilde{T}(r) \U(\Omega_1 + \Omega_2) \tilde{\Xi}_K(\Omega_1, \Omega_2, r_1, r_2),$$  \hspace{1cm} \text{(D20)}$$

where $\tilde{\Xi}_K(\Omega_1, \Omega_2, r_1, r_2)$ is as already given in Eq. (D10).

As for the time domain calculations, the susceptibilities for the condensate, non-condensate density and the non-condensate correlations are obtained by summing over the appropriate indices:

$$K^{(2)}_z(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{i=1}^{N} \left[ \tilde{T}(r) \left( \tilde{K}^{(2)}_I(\Omega_1, \Omega_2, r, r_1, r_2) + \tilde{K}^{(2)}_{II}(\Omega_1, \Omega_2, r, r_1, r_2) \right) \right],$$  \hspace{1cm} \text{(D21)}$$

$$K^{(2)}_\rho(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{i=2N+1}^{2N+N^2} \left[ \tilde{T}(r) \left( \tilde{K}^{(2)}_I(\Omega_1, \Omega_2, r, r_1, r_2) + \tilde{K}^{(2)}_{II}(\Omega_1, \Omega_2, r, r_1, r_2) \right) \right],$$  \hspace{1cm} \text{(D22)}$$

$$K^{(2)}_\kappa(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{i=2N+N^2+1}^{2N+2N^2} \left[ \tilde{T}(r) \left( \tilde{K}^{(2)}_I(\Omega_1, \Omega_2, r, r_1, r_2) + \tilde{K}^{(2)}_{II}(\Omega_1, \Omega_2, r, r_1, r_2) \right) \right],$$  \hspace{1cm} \text{(D23)}$$

3. Alternative form for the susceptibility

As before, expanding in the eigenstate basis $\tilde{\xi}_\nu$ which we introduced in Eq. (C33), we may write the susceptibility in a more useful form. The result is Eqs. (D21) - (D23) but with

$$\tilde{K}^{(2)}_I(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{\nu=1}^{2N(2N+1)} \mathcal{K}^{(2)}_I(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) \tilde{\xi}_\nu,$$  \hspace{1cm} \text{(D24)}$$

$$\tilde{K}^{(2)}_{II}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = \sum_{\nu=1}^{2N(2N+1)} \mathcal{K}^{(2)}_{II}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) \tilde{\xi}_\nu,$$  \hspace{1cm} \text{(D25)}$$

where

$$\mathcal{K}^{(2)}_{I,\nu}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_1, r_2) = -\frac{1}{4\pi^2} \sum_{\nu' \nu''=1}^{2N(2N+1)} \frac{\eta_{\nu}(r_1) \eta_{\nu'}(r_2) \mu_{\nu''}}{(\Omega_1 + \Omega_2 - \omega_{\nu'} + i\epsilon)(\Omega_1 - \omega_{\nu} + i\epsilon)(\Omega_2 - \omega_{\nu''} + i\epsilon)}.$$  \hspace{1cm} \text{(D26)}$$

Since no additional information is gained by listing all the terms in $\mathcal{K}^{(2)}_{II,\nu}(-\Omega_1 - \Omega_2; \Omega_1, \Omega_2, r, r_2)$, we simply note that, in the eigenstate basis, the typical term in $\mathcal{K}^{(2)}_{II,\nu}$ has the structure:

$$\sum_{\nu' \nu''=1}^{2N(2N+1)} \frac{\eta_{\nu}(r_1) \eta_{\nu'}(r_2) \mu_{\nu''}}{(\Omega_1 + \Omega_2 - \omega_{\nu'} + i\epsilon)(\Omega_1 - \omega_{\nu} + i\epsilon)(\Omega_2 - \omega_{\nu''} + i\epsilon)}.$$  \hspace{1cm} \text{(D27)}$$

as to be expected from Eqs. (D10) and (D20). $\mu_{\nu}$, and $\eta_{\nu}$ are as given in Eq. (C33).
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Fig. 1: Natural log of linear susceptibility $K^{(1)}(\Omega, \mathbf{r}, \mathbf{r}_1)$ at $\mathbf{r} = \mathbf{r}_1 = 0$ vs. frequency. Top three panels – zero temperature, bottom three panels – finite temperature $10\hbar\omega/k$. The frequency $\Omega_1$ are given in units of the trap frequency.

Fig. 2: $|K^{(2)}(t, t_1, t_2, \mathbf{r}, \mathbf{r}_1, \mathbf{r}_2)|$ i.e. the absolute value of the second order response functions in the time domain with the time $t_2$ fixed at $t_2 = 0$. (a) $r_2 = 0$; (b) $r_2 = -5$. The plots are for zero temperature condensate at the short, intermediate and long times $t$ and $t_1$ written at the top of each column of figures. The top, middle, and bottom rows give the response function for the condensate, non-condensate density, and non-condensate correlation respectively. The diameter of the dashed circle represent the spatial extent of the trapped BEC. The position $x$ and $x'$ are given in harmonic oscillator length units.

Fig. 3: Same as in Fig. 2, but at finite temperature of $10\hbar\omega/k$. The position $x$ and $x'$ are given in harmonic oscillator length units.

Fig. 4: $|K^{(2)}(\Omega_1, \Omega_2, \mathbf{r}, \mathbf{r}_1, \mathbf{r}_2)|$ i.e. the absolute value of the second order response functions in the frequency domain with the variable $\mathbf{r}_2$ set at (a) $r_2 = 0$ and (b) $r_2 = -5$. The plots are for zero temperature condensate at the frequencies $\Omega_1$ and $\Omega_2$ given at the top of each column. Denoting “off-resonant” when the frequency does not match an eigen value of $\mathcal{L}^{(2)}$ and “on-resonance” when a frequency matches an eigen value, these frequencies are chosen such that $\Omega_1$, $\Omega_2$, and $\Omega_1 + \Omega_2$ are off-resonant ($\Omega_1 = 2.23$, $\Omega_2 = 1.55$); both $\Omega_1$ and $\Omega_2$ are on-resonance while $\Omega_1 + \Omega_2$ is off-resonant ($\Omega_1 = 2.2$, $\Omega_2 = 1.5$); and finally frequencies chosen so that $\Omega_1 + \Omega_2$ is on-resonance ($\Omega_1 = 0.7$, $\Omega_2 = 1.5$). The position $x$ and $x'$ are given in harmonic oscillator length units.

Fig. 5: Same as in Fig. 4, but at finite temperature of $10\hbar\omega/k$. At finite temperature, the resonant frequencies are shifted from the zero temperature counterpart so that the actual frequency combinations used are the following: $\Omega_1$, $\Omega_2$, and $\Omega_1 + \Omega_2$ off-resonant ($\Omega_1 = 2.43$, $\Omega_2 = 1.6$); both $\Omega_1$ and $\Omega_2$ on-resonance with $\Omega_1 + \Omega_2$ off-resonant ($\Omega_1 = 2.43$, $\Omega_2 = 1.5$); and finally $\Omega_1 + \Omega_2$ on-resonance ($\Omega_1 = 0.92$, $\Omega_2 = 1.5$). The position $x$ and $x'$ are given in harmonic oscillator length units.

Fig. 6: Left column: $|K^{(2)}(\Omega_1, \Omega_2)|$ i.e. the absolute value of the second order response functions in the frequency domain as a function of $\Omega_1$ and $\Omega_2$ with the position variables set at $\mathbf{r} = \mathbf{r}_1 = \mathbf{r}_2 = 0$. There are three large peaks that completely dominates the plot so that the remaining peaks are not represented in the plot. Center column: Scaled $|K^{(2)}(\Omega_1, \Omega_2)|$. The three largest peaks shown in the left column were scaled down to the same magnitude as other peaks in the plot. Right column: $\log |K^{(2)}(\Omega_1, \Omega_2)|$ i.e. logarithm of the response functions presented in the left column to help visualize the large variation in the magnitude of $|K^{(2)}(\Omega_1, \Omega_2)|$. The top, middle, and bottom rows give the response function for the condensate, non-condensate density, and non-condensate correlation respectively. The frequency $\Omega_1$ and $\Omega_2$ are given in units of the trap frequency.
Fig. 7: Same as in Fig. 6, but at finite temperature of $10\hbar \omega / k$. The frequency $\Omega_1$ and $\Omega_2$ are given in units of the trap frequency.
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