Resonant Light Scattering to Measure BEC-Pairing

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ABSTRACT

We present a single-scattering formalism for incoherent resonant light scattering by dilute quantum gas systems such as the atomic-trap Bose-Einstein condensates. We show that resonant scattering gives access to more information than the dynamical structure factor, familiar from non-resonant scattering. In particular, we show that the detuning dependence of the incoherent scattering cross-section allows the direct determination of the BEC pairing density $\langle \psi \psi \rangle$, which is a broken symmetry and provides evidence that the condensate is not in a good number state.
The unusual properties of the atomic-trap Bose-Einstein condensates \[1\]–\[3\] make them prime examples of dilute many-body systems with highly interesting microscopic structures. Experimentally, the most convenient probe is resonant optical scattering. From the theoretical perspective, this poses an interesting problem – resonant scattering is a second-order process and cannot be described by means of the usual Van Hove theory \[4\]. Nevertheless, as Javaneinen pointed out \[5\] \[6\], a Van-Hove-like expression is recovered in the off-resonant limit. In this paper, we discuss a single scattering formalism that is valid for arbitrary values of the detuning.

Typically, the width $\gamma$ of the resonant transition is much larger than the trap frequency $\omega_T$ (twice the ground state energy of the trapping potential, $\hbar = 1$ in our units). Nevertheless, for some long-lived states, $\gamma$ can be comparable to the relevant excitation energies of the many-atom system, so that the scattering is fast on the scale of the excited atom motion, but not necessarily on the scale of the many-body dynamics. In that case, we find that resonant scattering gives access to more information about the many-body structure than the dynamical structure factor. In particular, for a dilute Bose condensate we are lead to the remarkable conclusion that resonant scattering allows a direct determination of the pairing density $\langle \psi \psi \rangle$, related to the Bose symmetry breaking.

It is instructive to recall the non-resonant scattering result. We adopt the convention that for an incident particle of momentum $k_{\text{in}}$ and frequency $\omega_{\text{in}}$, scattered into a state of momentum $k_{\text{out}}$ and frequency $\omega_{\text{out}}$, $q$ is the momentum transferred to the target system, $q = k_{\text{in}} - k_{\text{out}}$, and $\omega = \omega_{\text{in}} - \omega_{\text{out}}$. Van Hove showed that the differential cross section $d^2\sigma/d\Omega\,d\omega$, where $d\Omega$ is an infinitesimal solid angle, only depends on $q$ and $\omega$, and is equal to

$$\frac{d^2\sigma}{d\Omega\,d\omega} = |f(q)|^2 S(q, \omega),$$

(1)

where $f(q)$ is the scattering length describing the scattering of an incident particle by an individual target particle, and $S(q, \omega)$ is the dynamical structure factor of the many-body system. The structure factor is the Fourier-transform of the density-density correlation
function,
\[ S(q) = (2\pi)^{-1} \int d^3x \, d^4x' \exp[iq \cdot (x - x')] \langle \hat{\rho}(x')\hat{\rho}(x) \rangle, \tag{2} \]
where \( \langle \rangle \) denotes the thermally averaged expectation value over the initial states of the many-body target system, and where we introduced the four-vector notation, \( q \equiv (q, \omega) \), \( x \equiv (x, t) \), and \( q \cdot x = q \cdot x - \omega t \). In second quantization, the density operator is equal to:
\[ \hat{\rho}(x) = \hat{\psi}^\dagger(x)\hat{\psi}(x), \tag{3} \]
where \( \hat{\psi} \) and \( \hat{\psi}^\dagger \) are the annihilation and creation fields in the Heisenberg picture. The integration \( \int d^3x \) in (2) is over the three spatial components, the time component being fixed.

Unlike non-resonant scattering, resonant photon scattering involves two interactions – the atomic excitation while absorbing the incident photon, and the de-excitation accompanied by photon emission. In summing over scattering histories, we integrate over the photon absorption amplitude at \( x_1 \), the photon emission amplitude at \( x_2 \), and the amplitude for the excited atom to move from \( x_1 \) to \( x_2 \), which is the excited atom propagator \( G(x_2, x_1) \), where \( x_1 \neq x_2 \). To deal with this type of non-locality, it is useful to work in the Wigner-representation, where a two-point function, \( f(x_1, x_2) \) in coordinate representation, is represented by
\[ f_W(x; p) = (2\pi)^{-4} \int d^4r \ f(x + r/2, x - r/2) \exp[ip \cdot r]. \tag{4} \]
Similarly, we introduce the Wigner distribution operator:
\[ \hat{\rho}_W(x; p) = (2\pi)^{-4} \int d^4r \ \hat{\psi}^\dagger(x + r/2)\hat{\psi}(x - r/2) \exp[ip \cdot r], \tag{5} \]
the expectation value of which gives the Wigner distribution \([8, 9]\), the quantum analogue of the classical phase-space distribution function. A straightforward but somewhat lengthy derivation yields the following cross-section for light scattering involving an atomic transition of natural frequency \( \omega_0 \) and dipole moment \( \mathbf{d} \):
\[ \frac{d^2\sigma}{d\Omega \, d(\hbar \omega)} \left[ \epsilon_{\text{in}}, \epsilon_{\text{out}}; \Delta \right] = \frac{3\gamma}{4k} \left| (\hat{\epsilon}_{\text{in}} \cdot \hat{\mathbf{d}}) \ (\hat{\epsilon}_{\text{out}} \cdot \hat{\mathbf{d}}) \right|^2 \frac{1}{2\pi} \times \int d^3x \, d^4x' \, d^4p \, d^4p' \exp[iq \cdot (x - x')] G_W(x; p + k_+) \ G_W^*(x'; p' + k_+) \ \langle \hat{\rho}_W(x', p')\hat{\rho}_W(x, p) \rangle, \tag{6} \]
where $\Delta$ is the detuning of the incident photons, $\Delta = \omega_{\text{in}} - \omega_0$, $k_+$ is the average of the incident and outgoing photon momentum four-vector, $k_+ = (k_+, \omega_+) \equiv ([k_{\text{in}} + k_{\text{out}}]/2, [\omega_{\text{in}} + \omega_{\text{out}}]/2)$, $\hat{d} = d/|d|$ and $\hat{\epsilon}_{\text{in}}$ and $\hat{\epsilon}_{\text{out}}$ are the polarization directions of the incident and detected photons.

The propagator $G$ can be computed from the eigenstates of the effective potential $\tilde{V}(x)$ experienced by the excited atoms. Nevertheless, if the excited state lifetime is so short that the excited state experiences a change in $\tilde{V}$ small compared to the width $\gamma$, (i.e. $|F \cdot v|/\gamma^2 | \ll 1$, where $F$ is the force and $v$ the velocity of the excited atom), then $G_W$ is approximated accurately by the homogeneous propagator with $\tilde{V}(x)$ as position dependent shift:

$$G_W(x; p + k_+) = \frac{1}{p_0 + \omega_+ - [H_e(p + k_+, x) + \omega_0]}$$

$$= \frac{1}{\Delta - [H_e(p + k_+, x) - (p_0 - \omega/2)]},$$

where $p_0$ is the frequency-component of the momentum four-vector $p$, and $H_e$ is the self-energy of the excited atom, including the potential $\tilde{V}$ and the width $-i\gamma/2$. Expanding the propagator $G_W$ in powers of the inverse detuning, gives a $\Delta^{-1}$–expansion of the cross-section, with coefficients that are n-th order moments of the $[H_e(p + k_+, x) - (p_0 - \omega/2)]$–functions of the propagators (7) with respect to the $\langle \hat{\rho}(x'; p') \hat{\rho}(x; p) \rangle$–correlation function. At first sight, the appearance of $k_+$, instead of the momentum of the excited atom, might seem puzzling. The problem is resolved when we return to the ordinary coordinate representation using

$$\int d^4x \ d^4p \ \exp[iq \cdot x] \ F(p) \ \hat{\rho}_W(x; p) = \int d^4x \ \exp[iq \cdot x] \ \hat{\psi}^\dagger(x) \ \tilde{\mathcal{F}}(\hat{p} + q/2) \ \hat{\psi}(x)$$

$$= \int d^4x \ \exp[iq \cdot x] \ \hat{\psi}^\dagger(x) \ \tilde{\mathcal{F}}(\hat{p} - q/2) \ \hat{\psi}(x),$$

where $F(p)$ is an arbitrary function, and $\mathcal{F}(\hat{p})$ the operator obtained by replacing the $p$-vector in the expression for $F$, by the $p$-operator, $\hat{p}_j = \frac{1}{i} \partial/\partial x_j$, if $j = 1, 2$ or 3, and $\hat{p}_0 = i \partial/\partial t$. The right $\rightarrow$ and left $\leftarrow$ arrows indicate that the $p$-operator only acts upon the field operator immediately to the right or left. Since $q/2 + k_+ = k_{\text{in}}$, the $[H_e(p + k_+, x) - (p_0 - \omega/2)]$–functions give an operator $\mathcal{H}_e(k_{\text{in}} + \hat{p}; x) - i\partial/\partial t$. The time derivative $i\partial/\partial t$, acts on a ‘hole’
In this sense, $H_e - i \partial / \partial t$ describes the evolution of the excited atom-hole pair and we denote the operator $H_e - i \partial / \partial t$ by $H_{e-g}$. The cross-section is then equal to

$$d^2 \sigma [\hat{\epsilon}_{in}, \hat{\epsilon}_{out}; \Delta] \quad \frac{d^2 \sigma [\hat{\epsilon}_{in}, \hat{\epsilon}_{out}; \Delta]}{d\Omega d(\hbar \omega)} = \frac{1}{2\pi} \times \int d^3 x \, d^3 x' \exp [iq \cdot (x - x')] \langle \hat{\psi}^\dagger (x') \left[ \frac{1}{\Delta - \hat{H}_{e-g} (x')} \right] \hat{\psi} (x) \hat{\psi}^\dagger (x) \left[ \frac{1}{\Delta - \hat{H}_{e-g} (x')} \right] \hat{\psi} (x) \rangle .$$

For large detunings, we approximate the denominators in (9) by $\Delta$, giving the off-resonant limit reported by Javaneinen [5]:

$$\lim_{\Delta \to \infty} \frac{d^2 \sigma [\hat{\epsilon}_{in}, \hat{\epsilon}_{out}; \Delta]}{d\Omega d(\hbar \omega)} = \frac{1}{\Delta} \left( \frac{3\gamma}{4k} \right)^2 S(q, \omega) ,$$

where we recognize the off-resonant limit of the scattering length for optical resonant scattering from atoms, showing the similarity of (10) to the Van-Hove expression (1).

The general result (9) indicates how and when the off-resonant limit (10) breaks down. Especially for cold fermionic atom systems, it can be important to correctly include the effects of the Doppler shifts, contained in (9). For the atomic Bose condensates, the recoil energies can be of importance, as we show below.

A straightforward generalization of the customary detailed balance argument [10] for the structure factor shows that changing the sign of the energy transfer $\omega \to -\omega$ (while keeping $\Delta$ constant) similarly gives:

$$d^2 \sigma \left( \Delta; \omega \right) = \exp (\beta \omega) \, d^2 \sigma \left( \Delta - \omega; -\omega \right) ,$$

where $\beta$ is the inverse temperature. Verifying (11) experimentally (which does not require angular resolution) for the atomic-trap BEC systems can test the hypothesis that the observed condensate is in thermal equilibrium and provides a direct measurement of the temperature.

We now discuss scattering from a dilute homogeneous BEC at zero temperature. Since our concern is not with coherent scattering (observed at $\omega = 0$, $q = 0$), we calculate the incoherent part of the cross-section, $d^2 \sigma_{nc} / d\Omega d\omega$, subtracting $\langle \hat{\psi}^\dagger \hat{\psi} \rangle \langle \hat{\psi}^\dagger \hat{\psi} \rangle$ from $\langle \hat{\psi}^\dagger \hat{\psi} \hat{\psi}^\dagger \hat{\psi} \rangle$ in (11). We expand the $\hat{\psi}$-fields in plane wave states, $\hat{\psi} (x) = V^{-1/2} \sum_k a_k (t) \exp [i k \cdot x]$, $\hat{\psi}^\dagger(x) = V^{1/2} \sum_k \overline{a_k (t)} \exp [i k \cdot x]$ and $\hat{\psi}^\dagger(x) = V^{-1/2} \sum_k a_k (t) \exp [-i k \cdot x]$. This gives
\( V^{-1/2} \sum_k a_k^\dagger(t) \exp[-i \mathbf{k} \cdot \mathbf{x}] \), where \( V \) is the volume \((V \rightarrow \infty \text{ in the end})\), and make the mean-field Bogoliubov approximation, treating \( a_{k=0} \), \( a_{k=0}^\dagger \) as c-numbers, \( a_{k=0}, a_{k=0}^\dagger \rightarrow \sqrt{N_0} \), keeping terms up to order \( N_0 \). The result is

\[
\frac{d^2 \sigma_{nc}}{d\Omega \, d\omega}[\hat{\epsilon}_{in}, \hat{\epsilon}_{out}; \Delta] \approx N_0 \left| (3\gamma/4k)(\hat{\epsilon}_{in} \cdot \hat{\mathbf{d}})(\hat{\epsilon}_{out} \cdot \hat{\mathbf{d}}) \right|^2 \delta(\omega - \omega_q) \times
\]

\[
\left( \frac{1}{\tilde{\Delta} + i\gamma/2 - \omega_q} \right)^2 \left( a_{-q}^\dagger a_{-q} + \frac{1}{\tilde{\Delta} + i\gamma/2} \right)^2 \left( a_q a_q^\dagger \right) +
\]

\[
+ \left[ \frac{1}{\tilde{\Delta} - i\gamma/2 - \omega_q} \right] \left[ \frac{1}{\tilde{\Delta} + i\gamma/2} \right] \left( a_{-q}^\dagger a_q^\dagger \right)
\]

\[
+ \left[ \frac{1}{\tilde{\Delta} + i\gamma/2 - \omega_q} \right] \left[ \frac{1}{\tilde{\Delta} - i\gamma/2} \right] \left( a_q a_{-q} \right),
\]

(12)

where \( \tilde{\Delta} \) is the effective detuning, \( \tilde{\Delta} = \Delta - \bar{\omega}(|k|) - \bar{V} \), and \( \bar{\omega}(|k|) \) represents the kinetic energy of an excited atom with momentum equal to the resonant wave number. In addition to the peak at \( \omega = \omega_q \) represented by (12), the incoherent scattering spectrum at fixed scattering angle and detuning, has a continuous background, caused by \( \mathbf{k} \)-modes \((\mathbf{k} \neq 0)\) excited to \( \mathbf{k} + \mathbf{q} \) states. In most cases of interest, the ratio of the integrated background intensity to the peak intensity is of the order of the depletion \((N - N_0)/N_0\), where \( N \) is the total number of atoms.

The Bogoliubov approximation implies that in each process a particle is taken out of, or put into the condensate. This leaves only two possibilities if \( \mathbf{q} \) is the momentum transfer: a particle is excited from the condensate and ends up into a final state of momentum \(+\mathbf{q}\) (corresponding to the subscript \( \mathbf{q} \) in (12)), or a particle is taken from the initial state of momentum \(-\mathbf{q}\) and ends up in the condensate (corresponding to the \(-\mathbf{q}\)-subscripts). In the \(+\mathbf{q}\)-process, the excited atom has momentum \( \mathbf{k}_{in} \), in the \(-\mathbf{q}\) process, the excited atom has momentum \(-\mathbf{q} + \mathbf{k}_{in} = \mathbf{k}_{out} \). In either case, the excited atom has a center-of-mass energy that is \( \bar{\omega}(|k|) + \bar{V} \). On the other hand, the initial-state energy for the \( \mathbf{q} \) and \(-\mathbf{q}\) processes are different, so that the \( \mathcal{H}_{e-k} \)-operator gives \( \bar{\omega}(|k|) + \bar{V} - i\gamma/2 \) for the \(+\mathbf{q}\)-process and \( \bar{\omega}(|k|) + \bar{V} - \omega_q - i\gamma/2 \) for the \(-\mathbf{q}\) scattering. The \(-\mathbf{q}\) process leaves the condensate with an extra particle, whereas in the \(+\mathbf{q}\) process, a particle is removed from the condensate. In
the usual single particle picture, one could expect these processes to give orthogonal final states, thereby precluding any interference. Nonetheless, the condensate is expected to be in a coherent state, rather than a number state, and the $+q$ and $-q$ final states have a finite overlap proportional to $\langle a_qa_{-q} \rangle$. Conversely, detecting the interference of the $+q$ and $-q$ scattering events answers the question whether or not the condensate is in a good number state. With regards to this issue, which has received considerable attention in the recent literature [11]–[15], we note that the incoherent scattering scheme provides an alternative to the interfering condensate experiments.

To experimentally detect the interference, we note that the difference in recoil energy for the $+q$ and $-q$ scattering processes gives a distinct detuning dependence to the interference contribution in the cross section (12):

$$
\frac{d^2\sigma_{nc}}{d\Omega d\omega} [\hat{e}_{in}, \hat{e}_{out}; \Delta] \approx N_0 \left| (3\gamma/4k) (\hat{e}_{in} \cdot \hat{d}) (\hat{e}_{out} \cdot \hat{d}) \right|^2 \delta(\omega - \omega_q) \times
$$

$$
\frac{1}{(\Delta - \omega_q)^2 + (\gamma/2)^2} \left[ \langle a_{-q}^\dagger a_{-q} \rangle + \langle a_qa_{-q} \rangle \right] + \frac{1}{(\Delta)^2 + (\gamma/2)^2} \left[ 1 + \langle a_q^\dagger a_q \rangle + \langle a_qa_{-q} \rangle \right]
$$

$$
- \frac{\omega_q^2}{(\Delta - \omega_q)^2 + (\gamma/2)^2} \times \frac{1}{(\Delta)^2 + (\gamma/2)^2} \langle a_{q}a_{-q} \rangle,
$$

(13)

where we used that $\langle a_{-q}^\dagger a_{-q} \rangle = \langle a_q^\dagger a_q \rangle$, $\langle a_q^\dagger a_q \rangle = 1 + \langle a_q^\dagger a_q \rangle$ and $\langle a_{-q}^\dagger a_{q} \rangle = \langle a_qa_{-q} \rangle$. Thus, the detuning dependence of the peak intensity differs from the simple Lorentzian $\left[ (\Delta^2 + (\gamma/2)^2) \right]^{-1}$. The simplicity of the actual detuning dependence, which contains only two parameters, the occupation number $\langle a_q^\dagger a_q \rangle$ and pairing matrix element $\langle a_{-q}a_q \rangle$, suggests a simple fitting of the experimental curves to determine their values. In Fig.(1) we show the detuning dependence of the intensity in the peak of the incoherent scattering spectrum, for the special case that the scattering angle corresponds to a momentum transfer for which the excitation energy $\omega_q$ is equal to the chemical potential $\mu$ and $\mu$ is equal to the width $\gamma$. The full curve shows the actual intensity, whereas the dotted lines indicate the contributions proportional to the occupation number and the pairing matrix element. The occupation number and pairing matrix elements where calculated in the Bogoliubov approx-
imation at T=0 (the finite temperature generalization is straightforward, see for example [16]). Requiring the non-Lorentzian contribution to be measurable leads to the condition that $\omega_q$ is of the order of $\gamma$. The Bogoliubov theory at T=0 gives $\langle a_q a_{-q} \rangle = -\mu/2\omega_q$, yielding a magnitude of the non-Lorentzian term relative to the other contributions that is of the order of $\sim (\frac{\omega_q}{\gamma})(\frac{\mu}{\gamma^2})$. Therefore, the best signal is obtained for backscattering, $q = 2k$, $\omega_q \sim 4\omega_r$, where $\omega_r$ is the recoil energy, with a relative magnitude of $\sim 2 (\frac{\omega_r}{\gamma^2})(\frac{\mu}{\gamma^2})$.

Of course the atomic-trap system is not homogeneous. In fact, we cannot require the condensate to be too close to homogeneity because if the system is optically thick, the single scattering approximation breaks down. Nevertheless, it is possible to have an optically thin system for which $\mu \gg \omega_T$, in which case the Thomas-Fermi description is valid and the condensate [17], [18], as well as the fluctuations [19], [20] behave locally in the same manner as the homogeneous system. In a Thomas-Fermi description of the incoherent scattering, the single peak is 'broadened', giving a feature in the frequency interval from $\sqrt{(q^2/2m + \mu)^2 - \mu^2}$ to $q^2/2m$. Within this feature, the intensity of a frequency interval $\omega'$ to $\omega' + d\omega$ has information about the spatial region in which the local excitation with momentum $q$ has an excitation energy $\omega_q(r)$ in the $d\omega$ - interval, $\omega' < \omega_q(r) < \omega' + d\omega$. If the density is nearly constant in this spatial region, the detuning dependence of the intensity in the $d\omega$ - interval is described by the above theory.

In conclusion, we have shown that resonant light scattering can measure the pairing matrix elements of Bose-Einstein condensates. The finite value of the 'pairing' density, $\langle \psi^\dagger \psi \rangle$, is an example of a higher-order broken symmetry and is of fundamental interest. Measuring its value would constitute a detailed test of mean-field theories and finding a non-zero result would prove experimentally that the condensate is not in a number state.

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Figure 1: Plot of the detuning dependence of the peak intensity in the incoherent scattering spectrum. The calculation was performed in the Bogoliubov approximation at zero temperature for the special case that the momentum transfer corresponds to an excitation energy $\omega_q$ equal to the chemical potential $\mu$, and $\mu$ is equal to the excited state width $\gamma$. The full curve shows the actual intensity, the dotted line with negative values shows the contribution proportional to the pairing number $\langle a_q a_{-q} \rangle$ (which is negative). The dotted line with the positive values represents the contribution proportional to the occupation number $\langle a_q^{\dagger} a_q \rangle$. 
Incoherent Scattering Peak Intensity

Detuning in units of $\gamma/2$

Intensity (arbitrary units)