Collective excitations in superconductors: From Bardeen-Cooper-Schrieffer theory to Bose condensation

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We study the collective excitations of a system with local attractive interactions as a function of the coupling strength. The two-particle excitations are obtained in the random-phase approximation. For the strong-coupling limit we reproduce the excitation spectrum of a hard-core Bose gas, while in the weak-coupling limit we obtained a spectrum consistent with the BCS picture. The evolution of the excitation spectrum from the weak- to the strong-coupling limits is continuous. We briefly discuss the effect of these excitations in the thermodynamic properties of the system.

I. INTRODUCTION

The problem of the evolution from weak-coupling superconductors to the Bose condensation of composite bosons as a function of the attractive interaction strength has been addressed by many authors.[1-5] Consider a gas of fermions with an attractive interaction $U(r)$. If U is small, the usual BCS instability takes place. The ground state is well described by the BCS wave function, pairing is a collective effect, and the thermodynamics is dominated by pair-breaking excitations with an energy gap 2Δ. In the opposite limit, large-U bound states of single pairs can occur. The system then can be described as a gas of bosons which exhibits a Bose condensation. It has been shown that the ground state evolves continuously from one limit to the other. However, the excitations and consequently the thermodynamic properties are quite different in both limits.[3] There are two relevant lengths in the problem: the size of the pairs ξ_0, which in the weak-coupling limit is the BCS coherence length, and the average interparticle distance r_0. The limit $\xi_0/r_0 \gg 1$ corresponds to the BCS picture. For $\xi_0/r_0 \ll 1$ pairs do not overlap and the coherence of the ground state can be destroyed without breaking them. The intermediate case $\xi_0/r_0 \cong 1$ may be relevant in the context of high-T_c systems which have a low density of carriers and a very short coherence length.

A simple model which can be used to describe the crossover between these two regimes is the negative-U Hubbard model. In the following sections we study the two-particle collective excitations of this model for arbitrary values of the interaction U and different particle densities. In Sec. II we present the model and calculations, and Sec. III includes results and discussion.

II. MODEL AND CALCULATIONS

Our starting point is the negative-U Hubbard model, which is given by

$$H=-t\sum_{i,j}c_{i\sigma}^{\dagger}c_{j\sigma}-U\sum_{i}c_{i\uparrow}^{\dagger}c_{i\uparrow}c_{i\downarrow}^{\dagger}c_{i\downarrow}-\mu\sum_{i,\sigma}c_{i\sigma}^{\dagger}c_{i\sigma}\ , \tag{1}$$

where $c_{i\sigma}^{\dagger}$ creates a particle at site i with spin σ, t is the hopping integral, U is the strength of the attractive interaction, and μ is the chemical potential. In what follows we consider the average particle number per site n less than 1. In particular, we analyze the low-density limit. For $n \cong 1$ the superconducting state competes with a charge-density-wave state, an effect which is not discussed in this work.[6]

For the weak-coupling limit ($U/t \ll 1$), BCS theory describes correctly the ground state and thermodynamics of the system. There is, however, one type of excitation which is not included in the usual mean-field theory: Because of the lack of long-range interactions in Hamiltonian (1), there is a Goldstone mode with energy $\alpha\hbar v_F q$ (v_F is the Fermi velocity, α is a numerical constant of order 1, and q the crystal momentum), which corresponds to density oscillations. This type of excitation has been studied extensively in the past.[7-9] Long-range Coulomb interaction lifts this branch to the plasma frequency ω_p. It has been shown that in this weak-coupling limit plasmons are almost insensitive to the occurrence of superconductivity. As one may expect and we discuss below, in absence of long-range interactions the Goldstone mode retains a pure density-oscillation character only if its wavelength λ is much larger than the BCS coherent length. For $\lambda \cong \xi_0$ there is a strong mixing between density oscillations and pair-breaking excitations. One may ask how these modes evolve when the strength of the attractive interaction increases to reach the intermediate situation ($\xi_0/r_0 \cong 1$) or the strong-coupling regime. The latter has been also studied in some detail.[6] For the sake of completeness, we briefly summarize the results obtained for this regime.

For $U/t \gg 1$ particles are always coupled in pairs. An effective Hamiltonian up to order t^2/U can be obtained by projecting out states corresponding to unpaired particles. The effective Hamiltonian reads

$$H=J\sum_{\langle ij\rangle} S_i^z S_j^z-\tfrac{1}{2}(S_i^+S_j^-+S_i^-S_j^+) , \quad (2)$$

with $J=4t^2/U$.

In Hamiltonian (2) the spin-up and -down states at site i correspond, respectively, to an empty and doubly occupied state of the fermions in (1). The longitudinal antiferromagnetic term $S_i^zS_j^z$ describes a repulsion between nearest-neighbor doubly occupied sites, while the transverse ferromagnetic term accounts for the hopping of the composite boson (fermion pairs) through a virtual pair-breaking excitation. The particle density n is related to the total magnetization through $\langle S^z\rangle=(1-n)/2$. In spin-wave theory the ground state of Hamiltonian (2) corresponds to an ordered state of tilted spins plus zero-point fluctuations. The angle θ between the z direction and spins is fixed by the density $\cos\theta=1-n$. This ordered phase corresponds to a superfluid phase of the composite bosons. The superfluid order parameter is given by the magnetization in the x-y plane. The orientation of $\langle S^+\rangle$ is the phase of the superfluid order parameter. It is well known that the ground state in the mean-field approximation of Hamiltonian (2) corresponds to a BCS approximation of Hamiltonian (1). The low-energy excitations are clearly the sound waves of the superfluid phase of the hard-core Bose gas—magnons in the pseudospin language of Hamiltonian (2). The dispersion relation of this collective excitations is given by[10]

$$\omega_q^2=(\mathfrak{z}J)^2[1+(2\cos^2\theta-1)\gamma_q^2-2\cos^2\theta\gamma_q] , \quad (3)$$

where $\mathfrak{z}$ is the coordination number and $\gamma_k=(1/\mathfrak{z})\sum_\delta e^{ik\delta}$.

In this limit pair breakings are high-energy excitations with a characteristic energy of the order of U. The superfluid-normal phase transition is driven by the collective excitation described above.

Recently, it has been shown[11] that Hamiltonian (1) possesses a so-called "pseudospin" symmetry. By symmetry arguments one can show that there are at least two collective modes with infinite lifetime, one at the center and the other at the corner M of the Brillouin zone. The former is the Goldstone mode mentioned above, and the latter has an energy $-U-2\mu$. These results are exact for any sign of U, any filling of the band, and any dimension.

As we have already mentioned, the BCS ground state is a good approximation for both limits and it has been argued[3,12] that the evolution from weak- to strong-coupling superconductivity is smooth with the BCS wave function as a reasonable interpolation. Although the ground state for the negative-U Hubbard model has been studied in some detail, the low-energy excitation spectrum is unknown for the intermediate regime.

In what follows we calculate the collective excitations for arbitrary values of the crystal momentum q and ratio U/t. The excitation spectrum should reproduce the two limits discussed above and the exact results known for q at the center and at the corner of the Brillouin zone.

To calculate the collective excitations, we proceed in the following way: We use a Bogoliubov transformation to rewrite Hamiltonian (1) in the form $H=H_0+H'$ with

$$H_0=C+\sum_k E_k(\alpha_{1k}^\dagger\alpha_{1k}+\alpha_{2k}^\dagger\alpha_{2k}) , \quad (4)$$

where C is the ground-state energy $E_k=(\varepsilon_k^2+\Delta^2)^{1/2}$, ε_k is the kinetic energy measured from the chemical potential renormalized by the Hartree contribution $\tilde{\mu}=\mu+nU/2$, and Δ is the BCS order parameter. These last two quantities are calculated self-consistently according to the usual mean-field equations:[3]

$$\frac{N}{U}=\frac{1}{2}\sum_k\frac{1}{(\varepsilon_k^2+\Delta^2)^{1/2}} , \quad (5a)$$

$$n=\frac{1}{N}\sum_k\left[1-\frac{\varepsilon_k}{(\varepsilon_k^2+\Delta^2)^{1/2}}\right] , \quad (5b)$$

where N is the number of lattice sites.

The Hamiltonian H' describes the residual two-particle interactions and is given by

$$H'=-\frac{U}{N}\sum_{k,k',q}[A_{k,k',q}\alpha_{1k+q}^\dagger\alpha_{2k'+q}^\dagger\alpha_{2k'}\alpha_{1k}+B_{k,k',q}(\alpha_{1k+q}^\dagger\alpha_{1k'}^\dagger\alpha_{1k'+q}\alpha_{1k}+\alpha_{2k+q}^\dagger\alpha_{2k'}^\dagger\alpha_{2k'+q}\alpha_{2k})$$
$$+C_{k,k',q}\alpha_{1k+q}^\dagger\alpha_{2k'+q}^\dagger\alpha_{1k'}^\dagger\alpha_{2k}^\dagger+D_{k,k',q}\alpha_{1k+q}^\dagger\alpha_{2k'+q}^\dagger\alpha_{2k}^\dagger\alpha_{2k'}+F_{k,k',q}\alpha_{1k+q}^\dagger\alpha_{2k'+q}^\dagger\alpha_{1k'}^\dagger\alpha_{1k}+\text{H.c.}] . \quad (6)$$

The coefficients A, B, C, D, and F can be easily calculated and are given by the coherence factors u_k and v_k of the Bogoliubov transformation.

The excitations of the Hamiltonian H_0 correspond to the creation of two quasiparticles $\alpha_{1k}^\dagger\alpha_{2k+q}^\dagger$ and form a continuum with a gap $2\Delta(q)$. This gap is equal to the BCS gap 2Δ for $q<2k_F$ and increases for $q>2k_F$.

The residual interaction H' mixes these excitations and generates a low-energy collective mode in the gap. As H' does not conserve quasiparticle number, the excitation operator contains not only terms with two-quasiparticle creation operators, but terms with two-quasiparticle annihilation operators, thus taking into account ground-state correlations. Other possible terms such as $\alpha_k^\dagger\alpha_k$ do not appear in the simplest random-phase approximation (RPA). Then we propose an excitation operator

$$B_q^\dagger=\sum_k(a_k\alpha_{1k+q}^\dagger\alpha_{2k}^\dagger+b_k\alpha_{2k+q}\alpha_{1k}) , \quad (7)$$

where a_k and b_k are the solutions of the equations

$$(\omega-E_k-E_{k+q})a_k+\frac{1}{N}\sum_{k'}\gamma_{k,k',q}a_{k'}-\frac{1}{N}\sum_{k'}\delta_{k,k',q}b_{k'}=0 , \quad (8a)$$

$$(\omega+E_k+E_{k+q})b_k-\frac{1}{N}\sum_{k'}\gamma_{k,k',q}b_{k'}+\frac{1}{N}\sum_{k'}\delta_{k,k',q}a_{k'}=0 \ , \tag{8b}$$

where the coefficients $\gamma_{k,k',q}$ and $\delta_{k,k',q}$ are given by

$$\gamma_{k,k',q}=U(u_k U_{k+q}u_{k'}u_{k'+q}+v_k v_{k+q}v_{k'}v_{k'+q}+u_k v_{k+q}u_{k'+q}v_{k'}+u_{k+q}v_k u_{k'}v_{k'+q}) \ , \tag{9a}$$

$$\delta_{k,k',q}=U(u_{k+q}v_k u_{k'+q}v_{k'}+u_k v_{k+q}u_{k'}v_{k'+q}-v_k v_{k+q}u_{k'}u_{k'+q}+u_k u_{k+q}v_{k'}v_{k'+q}) \ . \tag{9b}$$

The integral equations (8) are solved numerically. In these equations we use the values of the gap parameter Δ and the chemical potential μ obtained self-consistently from Eq. (5). To minimize the computational work, we have considered a two-dimensional (2D) square lattice. However, in the present approximation we do not expect dimensional effects and the results are representative of the behavior of a 3D system.

III. RESULTS AND DISCUSSION

The energy ω_q of the collective mode associated with the operator $B_q^\dagger$ given in Eq. (7) is always smaller than the pair-breaking excitations with the same total momentum q. We first present the results corresponding to the large-U limit. The results for $U=15W$, where W is the half-bandwidth of the uncorrelated particles, and for different particle densities n are shown in Fig. 1. For the particle density n going to zero, the dispersion relation ω_q reproduces the one-particle spectrum. As the density increases, there is always a region for small q where ω_q is linear in q. For $q=(\pi,\pi)$—the zone boundary—the energy of the collective excitations decreases with increasing density, indicating the tendency of the system to form a charge-density wave. The dispersion relation in this limit is exactly the same as that given in Eq. (3), which was obtained by means of a canonical transformation of

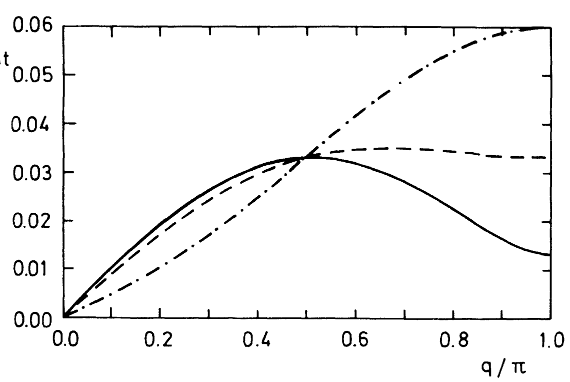

FIG. 1. Dispersion relations for the collective mode in the strong-coupling limit ($U=15W$, where W is the half-bandwidth of the uncorrelated fermions), for three different values of the band filling: $n=0.1$ (dot-dashed line), $n=0.5$ (dashed line), and $n=0.8$ (solid line).

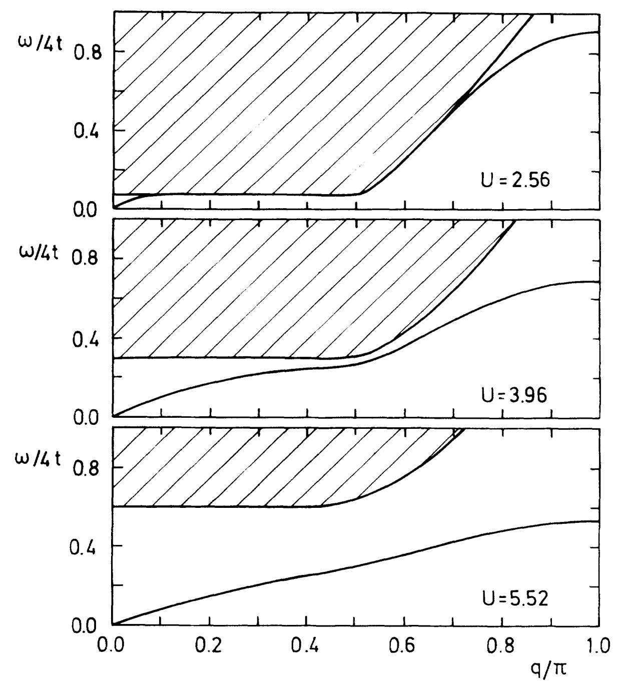

FIG. 2. Dispersion relations for the collective mode and two-particle continuum for $n=0.2$ and three values of the strength of the attractive interaction U.

the Hamiltonian in Eq. (1). Our RPA calculation then gives the correct result for the strong-coupling limit.

As U decreases, there are two effects: On the one hand, the total bandwidth of the collective excitations increases, and on the other hand, the energy of the pair-breaking excitations decreases. In Fig. 2 we present results of the two-particle excitation spectrum for different values of U corresponding to the weak-, intermediate-, and strong-coupling regimes. As U decreases, the velocity of the collective mode for q going to zero increases and in the weak-coupling limit it reaches the value $\hbar v_F(1-U\rho)/2$, v_F being the Fermi velocity and ρ the density of states at the Fermi level. This is a well-known result obtained by Anderson, although in the present case the numerical constant is different because we are working in a two-dimensional system. For small U this collective mode reaches the continuum for $q\cong 4\Delta/\hbar v_F$. This indicates that if the wavelength of the collective excitation is larger than the coherence length ξ_0, the sound wave can be propagated in the superconducting gas. Clearly, the existence of this Goldstone mode is due to the lack of long-range repulsion in the Hubbard model. For $q>4\Delta/\hbar v_F$ there is also a bound state with an energy which is very close to 2Δ except for q close to the zone boundary where there is a split of the two types of modes. For intermediate values of U, there is a qualitative change in the energy spectrum consisting of a split, for the whole range of q, of the energy of the two types of excitations. This change occurs for values of U and density such that $\xi_0\cong r_0$. This is precisely the Leggett criterion[1] for the crossover between a BCS regime and a Bose condensation of tightly bound pairs.

The crossover criterion corresponds to having the chemical potential just at the bottom of the band. This condition can be put in the form

$$U_c = \frac{2W}{\tanh^{-1}(1-n)} . \qquad (10)$$

For a fixed n the transition will be Bose like (BCS like) for $U > U_c$ ($U < U_c$).

As we mentioned above, because of the pseudospin symmetry of the Hubbard model, a state at the zone boundary $[Q=(\pi/a,\pi/a)]$ with an energy equal to $-U-2\mu$ should occur. The existence of this mode can be proved by defining the operator[11]

$$J_+ = \sum_j e^{iQR_j} c_{j\uparrow}^\dagger c_{j\downarrow}^\dagger , \qquad (11)$$

where R_j is the coordinate of site j. It can be easily shown that J_+ is an eigenoperator of H, satisfying the equation

$$[H,J_+] = (-U-2\mu)J_+ . \qquad (12)$$

Thus J_+ acting on the ground state creates an eigenstate of H with an excitation energy $-U-2\mu$. We have checked that the energy of the zone-boundary mode obtained in the RPA is given exactly by $-U-2\mu$ with the chemical potential μ given by the self-consistent equation (5). As expected, the RPA calculation preserves the pseudospin symmetry of the model Hamiltonian.

From the analysis of the symmetries of the Hubbard Hamiltonian, one can only conclude that there should be a Goldstone mode for $q \to 0$ and another mode for $q \to Q$. What our results indicate is that bound states exist for all values of q and the two modes obtained by symmetry arguments correspond to the same branch of excitations.

In order to check quantitatively our results, we have studied numerically a small cluster of 4×4 sites. We evaluate the ground-state energy and lowest excited states and compared the results with the RPA calculation for a cluster of the same size. In Fig. 3 we present this comparison for $q=Q$ and $Q/2$. As can be seen from the figure, the results are in excellent agreement.

These excitations will play an important role in both the spectral and thermodynamic properties of the system. These excitations are not coupled with transverse electromagnetic fields[13,14] and, in consequence, are not directly observable in optical transitions. However, other spectroscopies such as electron-energy-loss experiments could give a direct measure of these modes.

The effect of these modes on the thermodynamics may be very relevant: (i) In the large-U limit these excitations drive the superfluid-normal transition. (ii) In the weak-coupling limit the pair-breaking excitations are the ones responsible for the transition since we do not expect the collective mode to play an important role. Only very close to the zone center is there a low-energy excitation which differs in character from the pure pair-breaking excitation. In most of the Brillouin zone, the collective mode has an extremely small binding energy and the nature of the excitation is essentially of the pair-breaking type. (iii) In the intermediate regime both types of excitations will contribute to the thermodynamic properties. To visualize the characteristic energy of both types of excitations, we present in Fig. 4 the minimum pair-breaking energy and total bandwidth of the collective modes as a function of U.

The characteristic energies are of the same order of magnitude for U and n, satisfying the crossover criterion of Eq. (10).

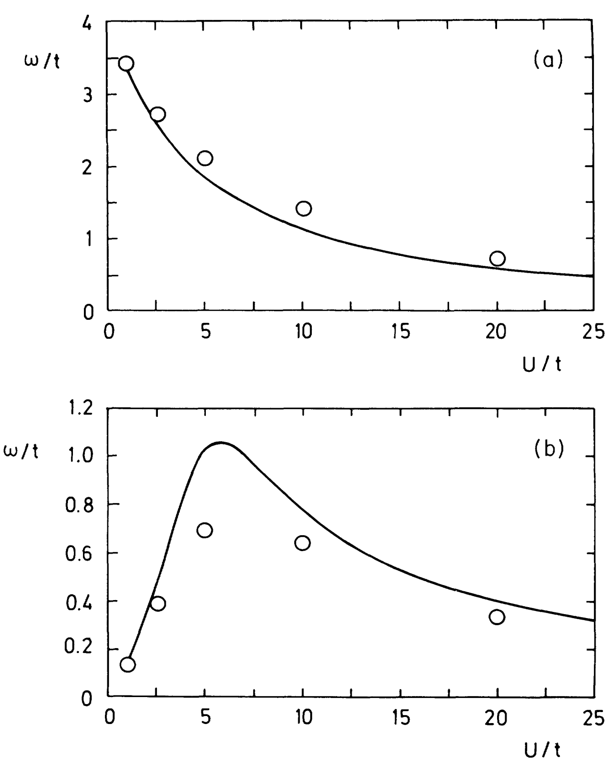

FIG. 3. Comparison between RPA results (solid line) and the exact diagonalization of 4×4 clusters with band filling $n=0.25$, for two values of the total crystal momentum: (a) $q=(\pi,\pi)$ and (b) $q=(\pi/2,\pi/2)$.

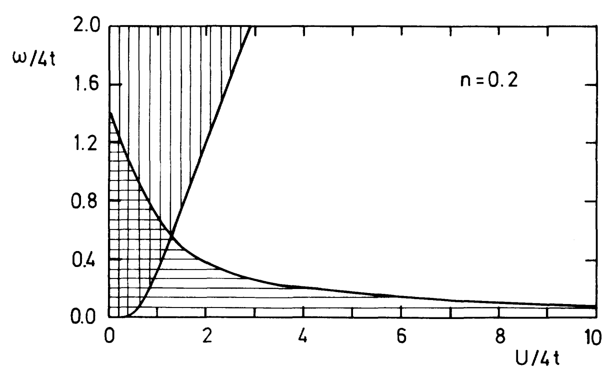

FIG. 4. Pair-breaking excitation energies (vertically shaded area) and total bandwidth of the collective mode (horizontally shaded area) as a function of the interaction strength U for $n=0.2$.

In summary, we have calculated the two-particle collective modes of the negative-U Hubbard model in the RPA. The calculation preserves the pseudospin symmetry of the Hubbard model. It reproduces correctly the weak- and strong-coupling limits. By comparing with exact results in a small cluster, we have shown that the approximation is qualitatively correct for all values of the interaction U. The collective modes evolve continuously from the weak- to strong-coupling limits. We have attempted to characterize the collective excitations of the intermediate regime, where the physics is richer and may be relevant in the context of the new high-T_c materials. The low-temperature properties of the model can be easily obtained from the knowledge of the excitation spectrum; however, in real systems the long-range Coulomb repulsion raises the Goldstone mode up to the plasma frequency. However, the long-range repulsion will not affect the short-wavelength excitations very much.

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[1] A. J. Leggett, in *Modern Trends in the Theory of Condensed Matter,* edited by A. Pekalski and J. Przystawa (Springer-Verlag, Berlin, 1980), p. 13.

[2] D. M. Eagles, Phys. Rev. **186**, 456 (1969).

[3] P. Nozières and S. Schmitt-Rink, J. Low Temp. Phys. **59**, 195 (1985).

[4] M. Randeria, J. Duan, and L. Shieh, Phys. Rev. B **41**, 327 (1990).

[5] R. T. Scalettar, E. Loh, J. Gubernatis, A. Moreo, S. White, D. Scalapino, R. Sugar, and E. Dagotto, Phys. Rev. Lett. **62**, 1407 (1990).

[6] S. Robaszkiewicz, R. Micnas, and K. A. Chao, Phys. Rev. B **23**, 1447 (1981).

[7] P. W. Anderson, Phys. Rev. **114**, 1002 (1959).

[8] N. N. Bogoliubov, V. V. Tolmachev, and D. V. Shirkov, *A New Method in the Theory of Superconductivity* (Consultants Bureau, New York, 1959), Chap. 4.

[9] G. Rickayzen, Phys. Rev. **115**, 795 (1959).

[10] A. Alexandrov and J. Ranninger, Phys. Rev. B **23**, 1796 (1981).

[11] S. C. Zhang, Phys. Rev. Lett. **65**, 120 (1990).

[12] The mean-field slave boson for the superconducting phase also gives a continuous evolution from the weak- to strong-coupling limit. The results are very similar to the BCS mean field with a small renormalization of the parameters for intermediate values of U: J. O. Sofo and C. A. Balseiro (unpublished).

[13] G. Rickayzen, *Theory of Superconductivity* (Interscience, New York, 1965).

[14] T. Tsuneto, Phys. Rev. **118**, 1029 (1960).