DLRO, ODLRO AND SUPERFLUIDITY

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ABSTRACT

A necessary condition for superfluidity is discussed in connection with the classification of the long-range orders, DLRO and ODLRO, and it is concluded that it can take place only in systems with ODLRO. Based on this consideration, the possibility of superfluidity in a system of Frenkel excitons is examined. It is shown that it cannot occur in the excitonic phase, but that it can occur in the Bose-condensed phase of high-density excitons as a transient phenomenon.

According to Yang,[1] the long-range orders taking place in liquid helium and superconductors are sometimes called as the off-diagonal long-range order (ODLRO). Characteristic features of these orders are that the Gauge symmetry of the system is broken in the ordered state, and that the order parameter is off-diagonal with respect to the number of particles. We may call the other type of orders as the diagonal long-range order (DLRO), where the symmetry broken in the ordered state is the rotational or translational symmetry in the configurational or spin space and the order parameter is diagonal with respect to the number of particles. It is usually believed that superfluidity can occur only in systems with ODLRO in the above sense.[2]

From a mathematical point of view, however, it seems rather artifitial to discriminate the Gauge symmetry from the other type of symmetries. As is well known, there are various cases where ODLRO is mathematically equivalent to DLRO. As an example, let us consider the quantum lattice-gas model of bosons introduced by Matsubara and

Matsuda.[3)] In this model, the Hamiltonian is given by

$$H = \frac{1}{2} \sum_{i>j} t_{ij}(a_i^+ a_j + a_j^+ a_i) + \sum_{i>j} U_{ij} n_i n_j - \mu \sum_j n_j , \tag{1}$$

$$n_j = a_j^+ a_j \quad .$$

Here a_j^+ and a_j, the creation and annihilation operators of bosons at the lattice site j, obey the following commutation relations:

$$\begin{aligned} &[a_i^+ , a_j^+]_- = [a_i , a_j]_- = [a_i , a_j^+]_- = 0 \qquad (i \neq j) \\ &[a_i , a_i]_+ = [a_i^+ , a_i^+]_+ = 0 , \qquad [a_i , a_i^+]_+ = 1 \quad . \end{aligned} \tag{2}$$

In eq.(1), the first term is the kinetic energy, the second term is the interaction between bosons, and μ denotes the chemical potential. The hard-core interaction between bosons is taken into account by the fermion-type commutation relation at the same lattice site. If we introduce the pseudospin operators by

$$\begin{aligned} &a_i \rightarrow S_i^x + iS_i^y \quad , \quad a_i^+ \rightarrow S_i^x - iS_i^y \\ &n_i \rightarrow \frac{1}{2} - S_i^z \quad , \end{aligned} \tag{3}$$

they obey the usual commutation relations of spin operators

$$[S_i^x , S_j^y] = iS_i^z \delta_{ij} \qquad etc.$$

Using these operators, we can rewrite the Hamiltonian (1) as

$$H = \sum_{i>j} [J_{ij}^z S_i^z S_j^z + J_{ij}(S_i^x S_j^x + S_i^y S_j^y)] - h \sum_i S_i^z , \tag{4}$$

where

$$J_{ij}^z = U_{ij} \quad , \quad J_{ij}^{\perp} = t_{ij} \quad , \quad h = \mu + \Sigma_j U_{ij} \quad .$$

In this example, we find an exact mathematical equivalence between a boson system and a spin system whose Hamiltonians are respectively given by eqs.(1) and (4). The mutual correspondence between the two systems is given in Table 1. Then it is rather self-evident

Table 1. Correspondence between a boson system and a spin system equivalent to it.

	boson system	spin system
symmetry	Gauge symmetry	Rotational symmetry around the z-axis
conserved quantity	The number of particles	The z-component of spins
order parameter	$\psi = \frac{1}{\sqrt{N}} \sum_j a_j$	$\vec{M}^{\perp} = \sum_j \vec{S}_j^{\perp}$
degeneracy of the ordered state	the phase of ψ	The direction of $\vec{M}^{\perp}$ in the x-y plane

that everything which occurs in the former system can occur in the latter, too. Since the superflow of particles occurs in the boson system, the superflow of the z-component of spins can occur in the spin system. This implies that, if we generalize the concept of superfluidity to general physical quantities, ODLRO is not a necessary condition for the occurrence of superfluidity.[4)] What is essential for it is the continuous degeneracy of the ordered state with respect to some degree of freedom as a result of the broken continuous symmetry.

When we consider real systems, however, we find an essential difference between a boson system and a spin system, or between the Gauge symmetry and the rotational symmetry in the spin space. In real spin systems, the rotational symmetry is only an approximate one and we always find some sort of anisotropy energy. If it is sufficiently small, we may neglect it when we discuss thermodynamical properties of the system. When we consider the direction of the spontaneous magnetization, however, it plays an essential role, how weak it may be. Due to the anisotropy energy, the symmetry of the spin system reduces to a discrete one, and the direction of the spontaneous magnetization is fixed to one of easy axes. Thus the superflow of spin angular momentum cannot take place in real spin systems. The Gauge symmetry is quite different from this. It is a universal symmetry of physical systems, and is not destroyed by any perturbation. This is the reason why we can find superfluidity only in systems with ODLRO in the original sense of Yang.

Based on these general considerations, we shall next discuss the possibility of superfluidity in an exciton system. For simplicity, we consider Frenkel excitons in a lattice of two-level atoms.[5] Assuming two atomic levels to be s- and p_z-levels, and taking only the dipole interaction between atoms, we get the Hamiltonian as

$$H = \sum_j \frac{E}{2}(a_{jp}^+ a_{jp} - a_{js}^+ a_{js}) + \sum_{i<j} \frac{p^2}{R_{ij}^3}\left(1 - \frac{3Z_{ij}^2}{R_{ij}^2}\right)(a_{ip}^+ a_{is} + a_{is}^+ a_{ip})(a_{jp}^+ a_{js} + a_{js}^+ a_{jp}) \quad , \tag{5}$$

where $a_{j\alpha}^+$ and $a_{j\alpha}$ (α=s,p) are respectively the creation and annihilation operators of electrons in the α-level of the atom j, E the atomic energy splitting, p the matrix element of an electric dipole moment, R_{ij} the distance between atoms i and j, and Z_{ij} its z-component. The Hamiltonian can be rewritten again by using the pseudospin operators defined by[6]

$$a_{jp}^+ a_{js} \rightarrow S_j^x + iS_j^y \quad , \quad a_{js}^+ a_{jp} \rightarrow S_j^x - iS_j^y \quad ,$$

$$\frac{1}{2}(a^+_{jp}a_{jp} - a^+_{js}a_{js}) \rightarrow S^z_j \quad ; \tag{6}$$

i.e. we have

$$H = - \sum_{i>j} J_{ij}S^x_iS^x_j - h \sum_i S^z_j \quad . \tag{7}$$

If we take into account more general interactions, then the Hamiltonian becomes

$$H = - \sum_{i>j} (J^x_{ij}S^x_iS^x_j + J^y_{ij}S^y_iS^y_j) - h \sum_i S^z_j \quad , \tag{8}$$

where $J^x_{ij} \neq J^y_{ij}$ in general. An essential difference of this system from the spin system described by the Hamiltonian (4) is the lack of the rotational symmetry around the z-axis.

If the exchange interaction is sufficiently strong compared with the magnetic field, spins are ordered in the x-y plane at low temperature. This is the so-called excitonic phase,[7] which corresponds to the order of electric dipoles in the real space. In contrast to the system with the Hamiltonian (4), in this case the direction of ordered spins is fixed depending on the relative magnitude of J^x_{ij} and J^y_{ij} . Therefore superfluidity cannot take place in the excitonic phase.

Next we consider the case where the magnetic field is strong and in the ground state all spins align in the z-direction. This corresponds to the usual insulating phase where all atoms are in the atomic s-level. Suppose the system is coherently excited to the state with $M_{x,y} \equiv \sum_j S^{x,y}_j \neq 0$. This is the Bose-condensed state of excitons discussed in detail by Hanamura and Haug.[8] Then in the molecular-field approximation, the magnetization obeys the equation of motion

$$\frac{dM_x}{dt} = (h - J_yM_z)M_y \ ,$$

$$\frac{dM_y}{dt} = -(h - J_xM_z)M_x \quad , \tag{9}$$

$$\frac{dM_z}{dt} = (J_y - J_x)M_x M_y \quad ,$$

where $J_\lambda = N^{-1} \sum_j J_{ij}^\lambda$. These equations can be solved exactly by using elliptic functions. Though the anisotropy $(J_x \neq J_y)$ prevents a free precession of the magnetization and M_z is not a constant of motion, the motion is still a periodic one around the z-axis. The direction of the magnetization, or the phase of condensed excitons, is not fixed here. The situations are quite different between two cases, an excitonic phase and a Bose-condensed phase of excitons, though the Hamiltonian has the same symmetry. In the present case, we may expect the superflow of excitons, if the condensed excitons have a spatially inhomogeneous phase.

It should be emphasized here, however, that this is true only within the molecular-field approximation. In eq.(9) the magnitude of the magnetization $M^2 = \sum_\lambda M_\lambda^2$ is a constant of motion, but it is not in the original Hamiltonian. It implies that M^2 decays gradually in the course of time by the coupling with fluctuations.

The situation may be more explicit, if we rewrite the Hamiltonian again by using the boson operators B_j^+ and B_j defined by

$$\begin{aligned} S_j^x + iS_j^y &= B_j^+(1 - n_j) \quad , \\ S_j^x - iS_j^y &= (1 - n_j)B_j \quad , \\ S_j^z &= n_j - \frac{1}{2} \quad , \end{aligned} \tag{10}$$

where $n_j = B_j^+ B_j$. Then the interaction contains terms which do not conserve the number of bosons. Among them, quadratic terms, *i.e.* terms proportional to B^+B^+ and BB, can be eliminated by the Bogoliubov transformation, but higher order terms cannot. Even if the system is isolated from other systems, *e.g.* photons and phonons, and the total energy of the system is conserved, the number of excitons is not conserved. Therefore in equilibrium excitons take the Planck distribution, not the Bose-Einstein distribution. It means that, if the system initially has the macroscopic number of excitons in one level, they decay gradually in the course of time, and that superfluidity can occur only as a transient phenomenon. Whether you call it as superfluidity or not depends on the time scale you are considering, and perhaps on your own taste.

REFERENCE

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2) W. Kohn and D. Sherrington: Rev. mod. Phys. 42 (1970), 1.
3) T. Matsubara and H. Matsuda: Progr. theor. Phys. 16 (1956), 569.
4) We may generalize the definition of ODLRO to general conserved quantities. For instance, the order of the spin system with the Hamiltonian (4) is off-diagonal with respect to the z-component of spins which is the conserved quantity in this system. In this sense ODLRO is a necessary condition for superfluidity.
5) The possibility of superfluidity in this system was discussed by S. Nakajima; preprint. See also Nakajima's paper in this proceedings.
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