SUPERFLUID AND EXCITONIC STATES

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ABSTRACT

The fundamental difference between DLRO and ODLRO in relation to superfluidity is demonstrated by comparing the pseudospin model of Frenkel excitons with the quantum lattice model of ^{4}He and assuming Bose condensation in both models. The argument is extended also to Wannier excitons.

Since the classification of Yang,[1] we have been accustomed to associate superfluidity with the off-diagonal long range order (ODLRO). A formal proof was indeed given by Kohn and Sherrington[2] on the lack of superfluidity in the case of the diagonal long range order (DLRO). An explicit calculation of the energy transport in the excitonic phase of semimetals (or semiconductors) was done by Zittartz,[3] who concluded the lack of superheatconductivity.

However, Hanamura and Haug[4] have argued recently that the Bose condensation of Wannier excitons may result in the superflow of energy in contrast to the conclusion of Zittartz, though one has no superfluidity of mass and charge in accordance with Kohn and Sherrington. Nagaoka[5] has pointed out, on the other hand, that in the case of DLRO characterized as a coherent state of a certain wave (charge, spin, strain *etc.*) it should always be possible to find some perturbation to destroy the "superflow" of the wave by clamping its phase. In the case of ODLRO, where the gauge symmetry of a basic matter field (*e.g.* the electron pair field in a superconductor) is broken, no other system than the superfluid itself can fix the phase[6] since any realistic perturbation should be gauge invariant.

The purpose of the present report is to point out that this fundamental difference between DLRO and ODLRO in relation to superfluidity may most clearly be seen by comparing the pseudospin model[7] of Frenkel excitons on the one hand with the quantum lattice model[8] of superfluid ^{4}He on the other. Thus, take a lattice of two-level atoms and let ground and excited levels of the j-th atom be represented by up and down states of the pseudospin σ_{jz}, respectively. We assume the Hamiltonian

$$H = -\sum_j E\sigma_{jz} - \frac{1}{2}\sum_{j\neq\ell}\sum J_{j\ell}\{(1+\lambda)\sigma_{jx}\sigma_{\ell x} + (1-\lambda)\sigma_{jy}\sigma_{\ell y}\}, \qquad (1)$$

where E is the atomic level separation, $J_{j\ell}$ are transfer matrix elements, and $0 \leqq \lambda \leqq 1$ is the symmetry breaking parameter. Following Hanamura,[9] we may replace E by E-μ if we wish to deal with quasi-steady states of a highly excited insulator, where μ is the chemical potential of the exciton.

When $\lambda=0$, (1) is invariant under the rotation of pseudospins around the third axis (the XY isotropy) and has the same form as the quantum lattice model of ^{4}He. In this model, E is the chemical potential of the He atom and the XY isotropy means the number conservation of atoms, *i.e.*, the gauge invariance in the usual sense. The symmetry should therefore be preserved whatever perturbation (*e. g.* the interaction with walls) we may add. In the case of excitons, on the other hand, we always find some processes, in which the number of excitons is not conserved, so that $\lambda \neq 0$ in general. For instance $\lambda=1$ if J arises from the electric dipole interaction.[7]

We now suppose that the Fourier transform $J(k)=\Sigma J_{j\ell}\exp[ik\cdot R_{j\ell}]$ is a positive maximum at k=0, where $R_{j\ell}=R_j-R_\ell$ is the relative lattice vector. Thus pseudospins must be almost all aligned in a certain direction at low temperature. In the harmonic approximation, the direction is determined by minimizing

$$\Omega = -\frac{1}{2}\left[E\cos\theta + \frac{1}{4}J(0)\sin^2\theta\,\{1+\lambda\cos 2\phi\}\right] . \qquad (2)$$

Here

$$\frac{1}{2} - <\sigma_{jz}> = \sin^2\frac{\theta}{2} \tag{3}$$

is the number of excitons per site and ϕ is the phase common to all the atomic polarizations.

For $E > E_c$, where $2E_c = (1+\lambda)J(0)$, we have the normal ground state represented by pseudospins all parallel to the third axis ($\theta=0$). The elementary excitation from this state, *i. e.*, the Frenkel exciton, is represented by the magnon of pseudospins. Strictly speaking, when $\lambda \neq 0$, the ground state contains some excitons[7] corresponding to the zero-point precession of pseudospins. We obtain $\phi=0$ by minimizing this zero-point energy.

For $E < E_c$, we have the excitonic state, which is represented by pseudospins inclined with $\theta = \cos^{-1}(E/E_c)$. From (3), we see then that the number of zero-point excitons with k=0 is macroscopic (Bose condensation of Frenkel excitons). We also see from (2) that the phase ϕ is fixed to zero once $\lambda > 0$, however small it may be.

In the harmonic approximation, it is not difficult to obtain the elementary excitation energy

$$\varepsilon_k = [\frac{1}{2}(1+\lambda)\ \{\lambda J(0) + \frac{1}{2}(1-\lambda)(J(0) - J(k))\} \times \{\sin^2\theta\ J(0) + \cos^2\theta\ (J(0) - J(k))\}\ ]^{\frac{1}{2}} \ . \tag{4}$$

It has the gap at k=0

$$\varepsilon_0 = [\frac{1}{2}\lambda(1+\lambda)\ \sin^2\theta\ ]^{\frac{1}{2}}\ J(0) \tag{5}$$

which vanishes for $\lambda=0$. From (4) we then obtain the phonon-like excitation corresponding to the phonon in superfluid ^{4}He. It reflects the degeneracy of the ground state energy which is independent of the phase ϕ when $\lambda = 0$.

This degeneracy leads to the "Josephson effect". Thus the flow of excitons through a narrow boundary between two bulk subsystems is given by

$$C = \Sigma\ J_{\ell r} \langle \sigma_{\ell x}\sigma_{ry} - \sigma_{\ell y}\sigma_{rx} \rangle\ , \tag{6}$$

where sites ℓ and r are on the left and right of the boundary, respectively. Suppose that the phase ϕ has a jump at the boundary and is uniform otherwise, so that $2\langle\sigma_{\ell x}\rangle = \sin\theta\cos\phi_{\ell}$, $2\langle\sigma_{\ell y}\rangle = \sin\theta\sin\phi_{\ell}$, *etc.* Ignoring the zero-point fluctuation, we obtain

$$C \simeq \sin^2\frac{\theta}{2}\ \Sigma J_{\ell r}\ \sin(\phi_r - \phi_{\ell})\ , \tag{7}$$

where the dependence on the sine of the phase difference is characteristic of the Josephson effect.[10] In order to satisfy the continuity equation, the phase ϕ should not be quite uniform, but have a small gradient in each subsystem. In fact, in the case of $\lambda=0$, the metastable state corresponding to a relative minimum of Ω is possible with the non-uniform phase $\phi_j = \mathbf{k}\cdot R_j$ and $\theta = \cos^{-1}(J(k)/2E)$. Then (6) gives the uniform superflow proportional to k. In the case of ^{4}He, this type of superflow is usually obtained by the use of the Galilean transformation.

These arguments can be extended to Wannier excitons. We assume the simple model of semimetals

$$H = \sum_k \psi_k^+ \xi_k \tau_z \psi_k - \frac{1}{2} g \Sigma \psi_k^+ \vec{\tau}\psi_{k+q} \cdot \psi_{\ell}^+ \vec{\tau}\psi_{\ell-q} + \Sigma e^{iq\cdot X_i} u\psi_k^+(1-\lambda+\lambda\tau_x)\psi_{k+q}\ , \tag{9}$$

where ξ_k is the one-particle energy measured from the Fermi level, g represents the short range interaction between electrons (up pseudospin) and holes (down pseudospin), ψ_k is the two-component destruction operator, and $\vec{\tau} = (2\sigma_x,\ 2\sigma_y)$, $\tau_z = 2\sigma_z$. We have assumed the XY isotropy of the interaction, through which the number of excitons is thus conserved. We have destroyed this symmetry by including in (9) the interband scattering due to impurities located at random sites X_i. Its importance relative to the intraband scattering is measured by $0 \leqq \lambda \leqq 1$.

When $\lambda=0$, the order parameter

$$\vec{\Delta} = -g \sum_k \langle \psi_k^+ \vec{\tau}\ \psi_k \rangle \tag{10}$$

may be oriented in any direction on the XY plane of pseudospin space. We again have the flexibility of the order parameter as regards the phase ϕ and this may result in superfluidity, which is described by a GL type equation[11] near the transition point. Such a possibility is neglected in the argument of Zittartz.[3]

Once $\lambda \neq 0$, on the other hand, the direction of (10) is fixed along the Y-axis. Take the case $\lambda=1$ for example. We apply the Hartree-Fock approximation to the interaction term in (9) and also the self-consistent Born approximation to the impurity scattering. The calculation is then the same as that of the superconductor with paramagnetic impurities,[12] whose spins are fixed all in one direction. Assuming constant density of states N_F near the Fermi level and small coupling constant gN_F, we obtain the self-consistency equation at temperature T

$$\vec{\Delta} = TgN_F \sum_{n=-\infty}^{+\infty} [\omega_n^2 + |\vec{\Sigma}_n|^2]^{-\frac{1}{2}} \vec{\Sigma}_n . \tag{11}$$

Here

$$\begin{aligned} (1+\Lambda_n)\,\Sigma_n^{(x)} &= \Delta_x \\ (1-\Lambda_n)\,\Sigma_n^{(y)} &= \Delta_y \\ (1-\Lambda_n)\,\omega_n &= (2n+1)\pi T \end{aligned} \tag{12}$$

and in terms of the life time τ due to the impurity scattering

$$\Lambda_n^{-1} = 2\tau\, [\,\omega_n^2 + |\vec{\Sigma}_n|^2\,]^{\frac{1}{2}} . \tag{13}$$

For simplicity let us restrict ourselves to the transition temperature and linearize[11] as

$$\begin{aligned} &\Delta_x \,[\, 1 - TgN_F \sum_n \,[(2n+1)\pi T + \frac{1}{2\tau}\,]^{-1}\,] = 0 , \\ &\Delta_y \,[\, 1 - TgN_F \sum_n \,[(2n+1)\pi T\,]^{-1}\,] = 0 . \end{aligned} \tag{14}$$

Hence the excitonic phase with $\Delta_x=0$, $\Delta_y \neq 0$ will appear at the same transition temperature as that of the pure system.

Finally we should mention that, in writing the interference factor as exp $[iq \cdot X_i]$ in (9), we have tacitly assumed a direct gap. In general, we should write the scattering part as

$$H_{imp} = \Sigma\ u\ e^{iq\cdot X_i}\ \psi_k^+ \{(1-\lambda) + \lambda \vec{h}_i \cdot \vec{\tau}\}\ \psi_{k+q}\ , \tag{15}$$

$$\vec{h}_i = (\cos QX_i,\ \sin QX_i)\ . \tag{16}$$

Here Q is the wave vector pointing from the bottom of the electron band to the top of the hole band. The problem is thus similar to the superconductor with randomly oriented impurity spins. After taking the average over X_i, no anisotropy in pseudospin space is left, so that the distinction between DLRO and ODLRO is by no means obvious. If we turn to the collective motion, instead of the individual excitations, however, we find that the phase of the excitonic condensate is again pinned down by impurity spins. Though we cannot go into the detail, the mechanism is similar to the one pointed out by Lee, Rice and Anderson[13)] in the case of CDW produced by the Peierls instability. In the case of superconductors with impurity spins, on the other hand, the phase of the Cooper pair remains free because of the gauge invariance.

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