Volume 2

LOW TEMPERATURE PHYSICS - LT 13

Quantum Crystals and Magnetism

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QUANTUM CRYSTALS

1

Plenary Topics

Quantum Crystals: Theory of the Phonon Spectrum

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Introduction

Quantum crystals are crystals with large zero-point motions, caused by a light mass and a weak interaction of the lattice particles. Among these are the solid phases of the quantum liquids ^{4}He and ^{3}He, molecular hydrogen, and solid neon. The existence of large zero-point motions can cause striking effects, e.g., the existence of a sizable nuclear exchange interaction in ^{3}He or a wavelike propagation of vacancies or isotopic impurities.

In this paper I explore the lattice dynamic aspects of quantum crystals. We are actually investigating strong anharmonicities which could be found in other crystals as well as near melting or near structural phase transitions. It turns out, however, that the anharmonicities in a quantum crystal can be much stronger than, for instance, those in one of the heavier rare gas crystals near melting.

A rather interesting aspect has come up quite recently. Inelastic neutron scattering experiments in both the solid[1] and the liquid[2] have revealed striking similarities, and we might ask the question: Does the solid show liquidlike behavior[3] or is it the other way around?[4]

Let me list the problems which have to be faced if a microscopic theory is intended. First, we note an expansion of the lattice due to the zero-point motions in much the same way as ordinary thermal expansion due to thermal vibrations. This expansion can actually be so large that even the next-neighbor distance would be beyond the inflection point of the interaction potential. If we try to start our theory with the harmonic approximation, we end up with imaginary frequencies—in other words, with an unstable crystal. This difficulty has, however, been overcome by the renormalized harmonic approximation[5] in which the harmonic coupling constants are averaged over the zero-point motions.

This brings another difficulty. The zero-point motions are actually large enough that there is a fair chance that two lattice particles can approach each other within the hard-core radius. This means short-range correlations have to be an essential part of our theory. There are actually several ways to accomplish this: for instance, Jastrow factors[6] or one or the other forms of a scattering matrix.[7] For the moment, however, we adopt a slightly more general point of view.[8]

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Renormalized Phonon Theory

Let me go through the essential steps of the theory without going into too much detail. Assume the crystal is under the influence of some external forces $\mathbf{f}_i$ representing the external pressure or some small disturbance, eventually time dependent. The equation of motion of the position operator $\mathbf{x}_i$ of any particular particle is then

$$-m\frac{\partial^2}{\partial t^2}\mathbf{x}_i = \sum_j{}' \nabla V(\mathbf{x}_i - \mathbf{x}_j) + \mathbf{f}_i \tag{1}$$

where m is the mass and $V(\mathbf{r})$ is the interaction of the lattice particles. Let

$$\mathbf{d}_i(t) = \langle \mathbf{x}_i(t) \rangle \quad . \tag{2}$$

be the expectation value of $\mathbf{x}_i$, i.e., the average position of particle i, in the presence of the external force, then

$$-m\frac{\partial^2}{\partial t^2}\mathbf{d}_i(t) = \mathbf{K}_i(t) + \mathbf{f}_i(t) \tag{3}$$

where

$$\mathbf{K}_i(t) = \sum_j{}' \langle \nabla V(\mathbf{x}_i - \mathbf{x}_j) \rangle = \sum_j{}' \int d^3r\, g_{ij}(\mathbf{r})\, \nabla V(\mathbf{r}) \tag{4}$$

is the average internal force on particle i due to the presence of the particles labeled by $j \neq i$. It has been expressed by the pair correlation function for a distinct pair of particles

$$g_{ij}(\mathbf{r}) = \langle \delta(\mathbf{r} - \mathbf{x}_i + \mathbf{x}_j) \rangle \tag{5}$$

where again the expectation value is in the presence of the external forces. Therefore, g_{ij} might be time dependent. In the absence of time-dependent external forces, the left-hand side of Eq. (3) actually vanishes and we recover an expression for the equation of state.

One quantity of primary interest is the displacement correlation function .

$$\begin{aligned}\mathbf{D}_{ij}(t,t') &= \delta\mathbf{d}_i(t)/\delta\mathbf{f}_j(t') \\ &= \langle \mathbf{x}_i(t)\,\mathbf{x}_j(t') \rangle - \langle \mathbf{x}_i(t) \rangle \langle \mathbf{x}_j(t') \rangle \end{aligned} \tag{6}$$

It describes how a disturbance $\delta\mathbf{f}_j(t')$ propagates through the crystal causing a change $\delta\mathbf{d}_i(t)$ of the expectation value of the position of particle i at time t. Since such a disturbance propagates as a phonon, at least in a harmonic crystal; this quantity is called the phonon propagator. It also contains information about equilibrium properties; for instance, $\mathbf{D}_{ii}(0,0)$ gives the mean square fluctuations of particle i around its equilibrium position described by $\mathbf{d}_i$.

Let me return for a moment to the pair correlation function $g_{ij}(\mathbf{r})$, which is one of the crucial quantities to calculate. We already have several pieces of information, for instance: (1) It has to be normalized to unity; (2) the first moment gives the average distance between particle i and j, which is also given by $\mathbf{d}_i - \mathbf{d}_j$; (3) its second moment gives the mean square fluctuations of this distance, which can also be expressed by $\mathbf{D}_{ij}(00)$; (4) its asymptotic form at small distances is that of the scattering problem of a pair of particles interacting with the true two-particle interaction. These pieces of information actually turn out to be sufficient to determine $g_{ij}(\mathbf{r})$ for given $\mathbf{d}_i$ and $\mathbf{D}_{ij}$.

Let me now come back to the displacement correlation function. Using Eqs. (6) and (3), we can find an equation of motion having in mind that $g_{ij}(\mathbf{r})$, and with it $\mathbf{K}_i(t)$, is a function of $\mathbf{d}_i(t)$ and $\mathbf{D}_{ij}(t,t')$

$$-m\frac{\partial^2}{\partial t^2}\mathbf{D}_{ij}(t,t') = \mathbf{1}\delta_{ij}\,\delta(t-t') + \sum_l \int d\tau\,\mathbf{M}_{il}(t\tau)\,\mathbf{D}_{lj}(\tau t') \tag{7}$$

where we have introduced the self-energy

$$\mathbf{M}_{ij}(tt') = \delta\mathbf{K}_i(t)/\delta\mathbf{d}_j(t')\big|_{\text{tot}} \tag{8}$$

We have for the moment considered $\mathbf{D}_{ij}(t,t')$ as a function of $\mathbf{d}_i(t)$ and the derivative has to be taken with respect to the explicit dependence of $g_{ij}(\mathbf{r})$ as well as with respect to the implicit dependence through the width given by $\mathbf{D}_{ij}$.

In physical terms the self-energy, a generalization of the dynamic matrix, is given by the change in the internal force on a particular particle, provided the equilibrium positions of some other particles are changed. The simplest assumption we can make about $g_{ij}(\mathbf{r})$ is that it is some function $g(\mathbf{r} - \mathbf{d}_i(t) + \mathbf{d}_j(t))$. Inserting this in Eqs. (4) and (8) and integrating by parts, the self-energy would simply be the second derivative of the interaction averaged over the pair distribution function. In the limit where $g(\mathbf{r}) \to \delta(\mathbf{r})$ we recover, obviously, the harmonic approximation. In the case of quantum crystals, however, this averaging yields real phonon frequencies.

In general, the functional dependence of $g_{ij}(\mathbf{r})$ on $\mathbf{d}_i(t)$ is more complicated, and even for fixed widths this means neglecting the implicit dependence through the $\mathbf{D}_{ij}$; it changes its shape for varying $\mathbf{d}_i(t)$ as shown in Fig. 1.

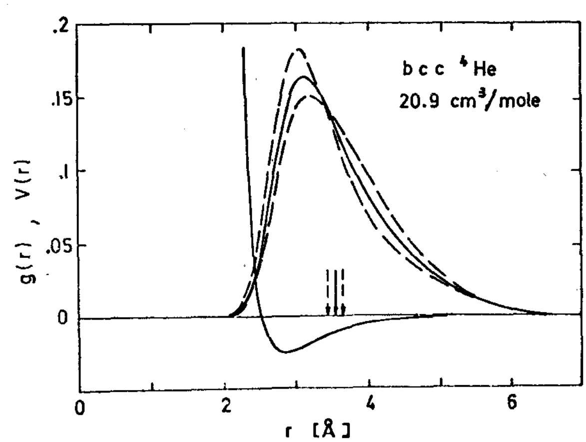

Fig. 1. Pair distribution function for three mean interparticle distances, indicated by arrows. Also shown is the interatomic potential for helium.

The way to proceed in a calculation would be to find a form for g_{ij} such that the conditions mentioned above are met and to calculate $g_{ij}(\mathbf{r})$ and $\mathbf{D}_{ij}(t)$ self-consistently.

Residual Anharmonicities

From this scheme, neglecting the dependence of $g_{ij}(\mathbf{r})$ on $\mathbf{D}_{ij}$ in calculating the self-energy, we obtain phonons without damping. Furthermore, the phonon, in this approximation, is a pure displacement motion.

If we include the dependence of $g_{ij}(\mathbf{r})$ on $\mathbf{D}_{ij}$ in lowest order, we obtain the bubble diagram (Fig. 2a) well known from ordinary anharmonic theory. The difference is, however, that the harmonic phonon frequencies in the intermediate lines are replaced by anharmonic ones, and the third-order coupling constants are replaced by renormalized vertices in very much the same way as the dynamic matrix was replaced by the renormalized harmonic vertex discussed above. As is well known, this diagram is responsible for phonon damping and for an additional anharmonic shift in the frequency.

We might take a slightly different point of view and say the third-order coupling constant represents a coupling between the one-phonon process (Fig. 2b) and the two-phonon process (Fig. 2c). This latter has a broad frequency distribution extending out to twice the maximum phonon frequency. This means that in the presence of this coupling the frequency distribution of the displacement response function now has not only a more or less sharp peak at the shifted phonon frequency, but in addition a tail ranging up to twice the maximum frequency and resembling the two-phonon frequency distribution. This is shown in Fig. 3 by the dashed lines.

The existence of this tail tells us that the true elementary excitation, represented by the sharp structure only, is no longer a pure displacement motion in the presence of the coupling. If we make a simple picture of a quantum solid where each particle has a Gaussian wave function near its lattice site, then a phonon in the absence of the coupling would be a collective oscillation of the rigid wave functions. In the presence of the coupling the wave functions also change their width in such a way that they narrow if neighboring particles move toward each other and widen if they move apart. This means that the actual elementary excitation is in general a coupled displacement and width fluctuation motion.

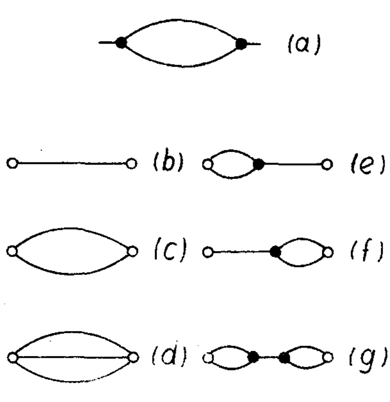

Fig. 2. (a) Bubble diagram contributing to phonon damping. (b–g) Diagrams for neutron scattering. (b) Bare single-phonon scattering $S_1^{(0)}(Q,\omega)$; (c–d) multiphonon processes $S_2^{(0)}(Q,\omega)$; (e–g) Interference terms $S_{\text{int}}(Q,\omega)$.

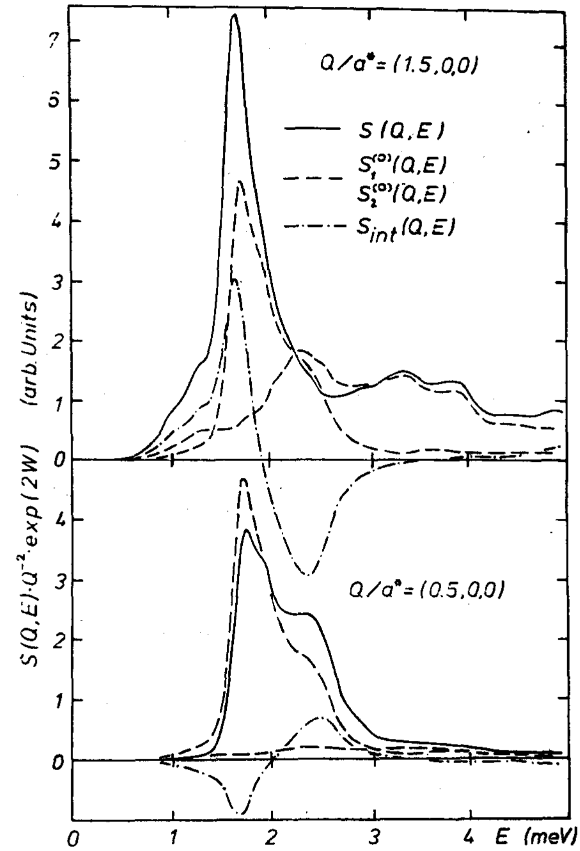

Fig. 3. Contributions to $S(Q, \omega)$ for two equivalent Q (see caption to Fig. 2.)

Neutron Scattering

As is well known,[10] the neutron scattering cross section in a crystal, in second Born approximation, is proportional to the dynamic scattering function

$$S(Q,\omega) = \frac{1}{2\pi N} \exp[-2W(Q)] \times \int_{-\infty}^{\infty} dt \exp(-i\omega t) \sum_{ij} \exp[i\mathbf{Q}\cdot(\mathbf{R}_i - \mathbf{R}_j)] \times \langle\langle \exp[i\mathbf{Q}\cdot\mathbf{u}_i(t)] \exp[-i\mathbf{Q}\cdot\mathbf{u}_j(0)] \rangle\rangle \tag{9}$$

where $\exp[-2W(Q)] = |\langle \exp(i\mathbf{Q}\cdot\mathbf{u}_i)\rangle|^2$ is the Debye–Waller factor. The double bracket stands for the cumulant of the corresponding expectation value plus one responsible for Bragg scattering. The usual way to evaluate the cumulant is