Eq. (13)]. More complicated correlations can be handled by taking recourse to the factorization property of averages of b's, as explained in Hecht (Ref. 16). The other two averages $\langle Q_m Q_e \rangle$ and $\langle P_m P_e \rangle$ provide us with a cross-check of our result that correlation functions with $\Gamma \neq 0$ vanish for infinitesimal coupling strength along the line.

[23]See the discussion in Ref. 1, immediately below Eq. (II 2.6). Incidentally, this equation is misprinted; it should read

$$\Phi(p_y) = [-(Ae^{ip_y} - 1)/(A - e^{ip_y})]^{1/2}.$$

Also, the operators σ of this reference are located on the y axis, as indicated by their coordinates $(0, k)$ and not on the x axis as stated in the text.

[24]See, for instance, N. I. Achieser, *Theory of Approximation* (Ungar, New York, 1956), p. 19.

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Fluctuations and Physical Properties of the Two-Dimensional Crystal Lattice

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The properties of finite, but large, two-dimensional crystal lattices are discussed in the light of the lack of long-range order. We confirm, with qualifications, the important basic result that the susceptibility diverges below a critical temperature. The details of our previous paper on Bragg peaks in scattering from the two-dimensional lattice are presented and the behavior of the dynamic structure factor $S(\vec{k}, \omega)$ about the peaks is analyzed. The lattice is shown to produce a Mössbauer peak with a non-Lorentzian line shape but with a Mössbauer strength of the same order of magnitude as that of the three-dimensional lattice. Finally, it is argued that finite phonon lifetimes would affect our results quantitatively but not qualitatively.

I. INTRODUCTION

The subject of long-range order in various one- and two-dimensional (2-d) systems has recently become a matter of great interest. There exist 2-d systems which possess long-range order, notably the Ising and probably the anisotropic Heisenberg models. On the other hand, we have many examples of 2-d systems for which long-range order can be rigorously shown not to exist,[1] for example, the isotropic Heisenberg model, a Bose condensate, electron pair superconductivity, and a crystalline[2] lattice. However, there is an increasing number of indications[3-5] that these last systems exhibit a variety of properties not too different from those characterising the three-dimensional (3-d) ordered analogs.

The reason for the interest in 2-d systems of the last type is that one would like to understand better the connection between mathematical long-range order and physical properties. Furthermore, it may be hoped that a 2-d geometry may be a good approximation for very thin layers and films and for materials with rod structures, when interest is focused on the motion perpendicular to the axis of alignment.

The 2-d crystal offers a particularly simple example. In the harmonic approximation it admits of an exact solution, and may serve, as shown by Jancovici,[6] as an example of a system with no long-range order that still has an "infinite" susceptibility. It has also been observed[4] that this nonordered structure gives rise to Bragg-like peaks in the x-ray structure factor, reminiscent of those obtained in ordered lattice structures. The reason for this effect is that the "divergence" in the mean-square fluctuation in the position of an atom, which leads to a vanishing order parameter, is caused by long-wavelength phonons, thus not affecting the short-range order. Related to this is the fact that the correlation function falls off slowly, as $1/r^{\alpha}$, and not exponentially.

The case of x-ray scattering is particularly simple because what is observed is the integral over all frequencies of the dynamic structure factor $S(\vec{k}, \omega)$, which is related to the equal-time correlation function. In the case of neutron scattering, however, one may have to discuss the detailed structure of $S(\vec{k}, \omega)$. A related problem is the "incoherent" or "self"-dynamic structure factor,[7] which determines[8] the Mössbauer line shape. When anharmonicity effects are considered, the problem becomes much more difficult. While the rigorous proofs of the nonexistence of long-range crystalline order have been carried over to the anharmonic case, we have only been able to present semiquantitative arguments (including sum rules) which indicate that our conclusions should remain valid also in the presence of weak phonon interactions.

In Sec. II we recapitulate the results for the susceptibility and the long-range behavior of the correlation function. Though these results are not new we stress that one may easily be led into significant errors by not being cautious enough with the procedure of taking the thermodynamic limit.

In Sec. III we discuss Bragg scattering in some detail. In Sec. IV we discuss the Mössbauer effect. Surprisingly we find that in the harmonic approximation a Mössbauer-like peak is possible with a shape differing from but reflecting the natural line shape. This happens in spite of the fact that the Debye-Waller factor is zero and is due to inelastic effects with small net energy transfer. The anharmonic problem is extremely delicate for the Mössbauer effect,[9] and we present only a rough argument indicating that our above result is preserved. It should be noted that in Sec. III stronger statements are made about anharmonicity effects in the coherent $S(\vec{k}, \omega)$, using sum rules and the "de Gennes narrowing effect." Most mathematical details are dealt with in Appendixes A and B.

II. SUSCEPTIBILITY AND LONG-RANGE ORDER

The intensive order parameter of a crystal lattice is given by the thermal average of the Fourier transform of the particle density,

$$\langle \rho_{\vec{k}} \rangle \equiv \langle (1/N) \textstyle\sum_n e^{i\vec{k}\cdot\vec{r}_n} \rangle \quad , \tag{2.1}$$

where $\vec{k}$ is a reciprocal-lattice vector, say $\vec{G}$, N is the number of atoms in the crystal, and $\vec{r}_n$ is the position vector of the nth atom. We can write

$$\vec{r}_n = \vec{R}_n + \vec{X}_n \quad ,$$

where $\vec{X}_n$ is the displacement from the equilibrium position $\vec{R}_n$ of the nth atom. Then,

$$\langle \rho_{\vec{k}} \rangle = F(\vec{k}) \langle e^{i\vec{k}\cdot\vec{X}_n} \rangle \quad , \tag{2.2}$$

where

$$F(\vec{k}) \equiv (1/N) \textstyle\sum_n e^{i\vec{k}\cdot\vec{R}_n} \tag{2.3}$$

has sharp peaks at $\vec{k} = \vec{G}$. For a harmonic lattice[10]

$$\langle e^{i\vec{k}\cdot\vec{X}_n} \rangle = e^{-\langle(\vec{k}\cdot\vec{X}_n)^2\rangle/2} = e^{-W} \quad , \tag{2.4}$$

where e^{-2W} is the Debye-Waller factor. W is well known to diverge for any 2-d lattice and thus there is no infinite-range order in infinite 2-d lattices. This is easily seen for the Debye lattice, when

$$2W = \frac{\hbar k^2}{8\pi m G^2} \int_0^{\omega_D} d\omega \coth\left(\frac{\hbar\omega}{2T}\right) , \tag{2.5}$$

where ω_D is the Debye frequency, m the atomic mass, c the sound velocity, and we have set Boltzmann's constant equal to unity. The integral diverges logarithmically as $\omega \to 0$, a property independent of the Debye approximation. For simplicity, in the remainder of this paper, we will refer to the classical 2-d lattice. Quantum effects exist once $T \lesssim \omega_D$, but they will have a crucial effect only for $T \lesssim \omega_D / N^{1/2}$. In the classical limit,

$$2W = (2T/T_k) \textstyle\int_0^{\omega_D} d\omega \, (1/\omega) \quad , \tag{2.6}$$

where $T_k = 4\pi mc^2 / vk^2$ and v is the unit-cell area.

A few years ago, Jancovici[6] pointed out that while there is no infinite range order in the lattice, the susceptibility χ_k diverges when $k = G$ and the temperature is less than T_G. This was very interesting for the reason that physicists have often accepted a singular susceptibility as an indication that the system has an instability which is dealt with by a change of state. Furthermore, numerical studies of the 2-d isotropic Heisenberg ferromagnet indicated[3] that its susceptibility would diverge at some nonzero temperature, whereas it can be rigorously shown[1] that no ferromagnetic state exists. Jancovici's result seemed to lend credence to these results.

The susceptibility $\chi_{\vec{k}}$ measures the linear response of the lattice to an external potential $\phi(\vec{r})$, the interaction energy being[11]

$$E_{\rm int} = -N \rho_{\vec{k}} \phi_{\vec{k}} \quad , \tag{2.7}$$

when $\phi(\vec{r}) = \phi_{\vec{k}} e^{i\vec{k}\cdot\vec{r}}$. Then, in the classical limit,

$$\chi_{\vec{k}} = (N/T) \left[\langle \rho_{\vec{k}}^2 \rangle - \langle \rho_{\vec{k}} \rangle^2 \right] \quad . \tag{2.8}$$

We thus see that we should consider a large finite crystal and carefully let $N \to \infty$.

It was pointed out recently[12] that for a finite crystal the logarithmic divergence of $2W$ is not very serious. In fact, there is a natural long-wavelength (and hence low-frequency) cutoff in the integral, namely, the linear dimension of the sample. Thus, with a cutoff frequency

$$\omega_c = \omega_D (\alpha/N)^{1/2} \tag{2.9}$$

where α is a constant of order unity,

$$2W = (T/T_k) \ln(N/\alpha) \quad , \tag{2.10}$$

so that

$$N \langle \rho_{\vec{G}} \rangle^2 = \alpha^{T/T_G} N^{1-T/T_G} \quad . \tag{2.11}$$

This is quite significant since, as Jancovici showed (see Appendix A), when $T < T_G$,

$$N\langle \rho_{\vec{G}}^2 \rangle \propto N^{1-T/T_G} + \text{const of order unity} \quad , \quad (2.12)$$

and thus the term $N\langle \rho_{\vec{G}} \rangle^2$ cannot be neglected in (2.8). In fact, when $T < T_G$,

$$N\langle \rho_{\vec{G}}^2 \rangle = \frac{\beta^{T/T_G} N^{1-T/T_G}}{1 - T/T_G} + \text{const of order unity} \quad , \quad (2.13)$$

where β is a constant of order unity. Since $\chi_{\vec{G}}$ is positive definite, we must have

$$\beta \geq \alpha \quad .$$

(Both α and β depend upon the crystal symmetry, shape, and boundary conditions.)

It is interesting to note that $\chi_{\vec{G}}$ also diverges in a 3-d lattice, which has infinite-range order and no instability. In particular (see Appendix A),

$$\chi_G \approx (T/T_G) N^{2/3} \quad . \quad (2.14)$$

In neither case [Eqs. (2.13) or (2.14)] does χ_G diverge as fast as N.

Jancovici's conclusion remains valid, namely, that

$$\chi_{\vec{G}} \to \infty \qquad \text{as } N \to \infty, \; T < T_G$$

$$\chi_{\vec{G}} \Big|_{N=\infty} \to \frac{\text{const of order unity}}{T - T_G} \quad \text{as } T \to T_G^+ \quad . \quad (2.15)$$

Thus the 2-d crystal is an example of a system whose susceptibility diverges without an instability occurring.

III. COHERENT SCATTERING AND BRAGG PEAKS

The cross section for coherent scattering of particles by a many-body system, with momentum transfer $\hbar\vec{k}$ and energy transfer $\hbar\omega$ to the system, is well known to depend upon the system only through the dynamical structure factor $S(\vec{k}, \omega)$ given by

$$S(\vec{k}, \omega) = \int_{-\infty}^{\infty} (dt/2\pi) e^{-i\omega t} I(\vec{k}, t) \quad , \quad (3.1)$$

where $I(\vec{k}, t)$ is the dynamical correlation function of the density fluctuation $\rho_{\vec{k}}(t)$:

$$I(\vec{k}, t) \equiv N \langle \rho_{\vec{k}}(t) \rho_{\vec{k}}(0) \rangle \quad . \quad (3.2)$$

We have already discussed two limiting behaviors of $I(\vec{k}, t)$,

$$I(\vec{k}) \equiv I(\vec{k}, t=0) = N \langle \rho_{\vec{k}}^2 \rangle \quad , \quad (3.3)$$

referred to as the "structure factor," and

$$I(\vec{k}, t \to \infty) = N \langle \rho_{\vec{k}} \rangle^2 \quad . \quad (3.4)$$

We can express $S(\vec{k}, \omega)$ as

$$S(\vec{k}, \omega) = N \langle \rho_{\vec{k}} \rangle^2 \, \delta(\omega) + S_R(\vec{k}, \omega) \quad , \quad (3.5)$$

where $\delta(\omega)$ is the Dirac δ function and $S_R(\vec{k}, \omega)$ will be shown to be well behaved (for finite N) but sharply peaked at $\omega = 0$.

In the case of typical x-ray scattering experiments one measures $I(\vec{k})$,[1] which can be expressed in terms of $S(\vec{k}\omega)$,

$$I(\vec{k}) = N \langle \rho_{\vec{k}}^2 \rangle = \int_{-\infty}^{\infty} d\omega \, S(\vec{k}, \omega) \quad . \quad (3.6)$$

Thus [cf. Eq. (2.13)], there will be a Bragg peak at $\vec{k} = \vec{G}$ when $T < T_G$. In Appendix B we derive the following behavior of $I(\vec{k})$ about a Bragg peak. With L the linear size of the lattice and

$$\vec{k} = \vec{G} + \vec{\kappa} \quad , \qquad \kappa \ll 1/v^{1/2}$$

$$\frac{I(\vec{k})}{I(\vec{G})} \approx 1 - \frac{1}{4} \frac{T}{T_G} \left(1 - \frac{T}{T_G}\right) \kappa^2 L^2, \qquad \kappa L \ll 1$$

$$\left.\begin{array}{ll} \approx \dfrac{1 - T/T_G}{(\kappa L)^{2(1-T/T_G)}} \;, & T/T_G > \frac{1}{4} \\[2ex] \approx \dfrac{\sin(\kappa L - \frac{1}{4}\pi)}{(\kappa L)^{3/2}} \;, & T/T_G < \frac{1}{4} \end{array}\right\} \; 1 \ll \kappa L \ll N^{1/2} \quad . \quad (3.7)$$

The oscillations which appear when $T/T_G < \frac{1}{4}$ are analogous to those appearing in the 3-d lattice.

Suppose now that neutrons are scattered off the crystal and that only energy transfers corresponding to the frequencies

$$|\omega| < \Delta$$

are detected. Then one measures

$$I_\Delta(\vec{k}) = \int_{-\Delta}^{\Delta} d\omega \, S(\vec{k}, \omega) \quad . \quad (3.8)$$

We show in Appendix B that as long as

$$\Delta \gg c/L \quad ,$$

we have

$$I_\Delta(\vec{k}) \approx I(\vec{k}) \quad . \quad (3.9)$$

Thus $S(\vec{k}, \omega)$ has a sharp peak about $\omega = 0$ of width on the order of c/L. Typically, we have $\Delta \approx 10^{11}$ $\text{sec}^{-1} \approx 10^{-4}$ eV, $c \approx 10^4$ cm/sec, and $L \approx 1$ cm, in which case

$$L\Delta/c \approx 10^7 \quad .$$

Nevertheless, suppose $\Delta \ll c/L$, then Bragg peaks would still be observed,

$$I_\Delta(\vec{k}) \to I(\vec{k}, t \to \infty) = N \langle \rho_{\vec{k}} \rangle^2 = F(\vec{k})^2 \, \alpha^{T/T_k} N^{1-T/T_k} \quad \text{as } \Delta \to 0 \quad . \quad (3.10)$$

We should like to point out that this type of behavior is very general and will also occur when anharmonicity is present provided only that it does not effect the peaks in $I(\vec{k})$. The reason is that the narrowing of $S(\vec{G}, \omega)$ follows from exact sum rules. In fact, it is an extreme manifestation of the well-known de Gennes narrowing effect.[13] As noted by de Gennes, the normalized second moment of (the coherent) $S(\vec{k}, \omega)$ is given by

$$\int \omega^2 S(\vec{k}, \omega)\, d\omega \Big/ \int S(\vec{k}, \omega)\, d\omega = k^2 T/mI(\vec{k}) \qquad (3.11)$$

in the classical approximation. Further details of the system appear only in the higher moments. For $\vec{k}$ near a peak of $I(\vec{k})$ the normalized second moment will be greatly reduced. In our case, assuming that the static $I(\vec{k})$ has a peak with a height of order N for $T \ll T_G$ (Eq. 2.13) (this "static" feature should not depend strongly on anharmonicity), we find that the normalized second moment of $S(\vec{G}, \omega)$ is of the order of N^{-1} and the typical width is therefore of the order of $N^{-1/2}$. For finite T/T_G, the sum-rule argument gives, in general, a sharpening of $S(\vec{G}, \omega)$, but it is weaker than in the pure harmonic case, the width being of the order of $N^{-(1-T/T_G)/2}$. We note in passing that $S(\vec{G}, \omega)$ can have any large ω tail which will not affect the second moment [e.g., $S(\vec{G}, \omega) \propto \omega^{-n}$, $n > 3$ behavior]. This tail will also certainly not affect the zeroth moment $I(\vec{k})$, but the moments higher than the second may well be large.

IV. MÖSSBAUER SPECTRUM

In this section we calculate the Mössbauer line shape due to the emission of a γ-ray photon of momentum $\vec{Q}$ from an atom in a 2-d crystal. We will begin with the harmonic crystal and later discuss qualitatively the effects of anharmonicity and impurities.

The line shape is given by[8,10]

$$P(\omega) = \int_{-\infty}^{\infty} (dt/2\pi)\, e^{-i\omega t - \Gamma|t|}\, C(\vec{Q}, t) , \qquad (4.1)$$

where Γ is the natural linewidth of the photon (typically about 10^8 sec^{-1}) and

$$C(\vec{Q}, t) = \langle e^{i\vec{Q}\cdot\vec{X}(t)}\, e^{-i\vec{Q}\cdot\vec{X}(0)} \rangle , \qquad (4.2)$$

with $\vec{X}(t)$ the displacement vector of the emitting atom from its equilibrium position at time t and $\langle\;\rangle$ signifying a thermal average at temperature T.

Using the usual procedure for calculating such a thermal average,[10] we obtain for the infinite 2-d classical Debye model

$$C(\vec{Q}, t) = e^{-f(\vec{Q}, t)} , \qquad (4.3)$$

where

$$f(\vec{Q}, t) = (T/T'_Q) \int_0^{\omega_D} d\omega\, \omega\, (1 - \cos\omega t)/\omega^2$$

or

$$f(\vec{Q}, t) = (T/T'_Q)\, [\ln\gamma\omega_D t - \mathrm{ci}(\omega_D t)] . \qquad (4.4)$$

Above, ω_D is the Debye frequency, $\ln\gamma = 0.577\ldots$ is Euler's constant, $\mathrm{ci}(x)$ is the cosine-integral function,[14] and

$$T'_Q = 2\pi m c^2/vQ^2 . \qquad (4.5)$$

One should note that $T'_Q = \frac{1}{2} T_Q$ [cf. Eq. (2.6)] and that

$$T'_Q \approx (\hbar\omega_D)^2/R_0 ,$$

where R_0 is the recoil energy of the unbound atom. To see what the order of magnitude of T'_Q is, suppose R_0 is 10^{-3} eV and $\hbar\omega_D$ is 200 °K, then $T'_Q \sim 4000$ °K.

For large t, i.e., $\omega_D t \gg 1$,

$$\mathrm{ci}(\omega_D t) \sim \sin(\omega_D t)/\omega_D t ,$$

so that

$$C(\vec{Q}, t) \sim (\gamma\omega_D t)^{-T/T'_Q} .$$

Since ω_D is on the order of 10^{13} sec^{-1} and thus $\omega_D \gg \Gamma$, we can be in this long-time region even when $\Gamma t \gg 1$. We are led to consider two temperature regimes: When $T > T'_Q$, $P(\omega)$ is essentially independent of Γ and has a characteristic width on the order of ω_D. When $T < T'_Q$, $P(\omega)$ depends upon Γ for $\omega \lesssim \Gamma$. To see this, we will decompose $P(\omega)$ into a sum of two parts,

$$P(\omega) = P_M(\omega) + P_R(\omega) , \qquad (4.6)$$

where

$$P_M(\omega) = \int_{-\infty}^{\infty} (dt/2\pi)\, e^{-i\omega t - \Gamma|t|}/(\gamma\omega_D t)^{T/T'_Q}$$

or

$$P_M(\omega) = \frac{\Gamma[1 - T/T'_Q]}{\pi(\gamma\omega_D)^{T/T'_Q}}\, \mathrm{Re}\left(\frac{1}{\Gamma + i\omega}\right)^{1-T/T'_Q} . \qquad (4.7)$$

Here, $\Gamma[x]$ is the Γ function of x.[14] Thus $P_M(\omega)$ has a width Γ. It is easily seen that $P_R(\omega)$ has a width ω_D.

We get an idea of the strength of the Mössbauer peak by looking at

$$\Gamma P_M(\Gamma) \approx (\Gamma/\omega_D)^{T/T'_Q} = e^{-(T/T'_Q)\ln(\omega_D/\Gamma)} . \qquad (4.8)$$

This quantity should be compared with its analog in three dimensions, the Debye-Waller factor e^{-2W}, where[8–10]

$$2W = 6R_0 T/(\hbar\omega_D)^2 \approx T/T'_Q .$$

We thus see that except for numerical factors of the order of unity in the exponents, there is no significant difference between the Mössbauer strengths in two and three dimensions. What is most significant is the non-Lorentzian line shape in two dimensions. However, unless T/T'_Q is of order unity, this shape will hardly be differentiated from a Lorentzian,

$$P_M(\omega) \to \frac{\Gamma/\pi}{\omega^2+\Gamma^2} \quad \text{as } T/T_Q' \to 0 \; . \tag{4.9}$$

In three dimensions, the Mössbauer effect is a zero-phonon process, wherein the Mössbauer peak has the nuclear line shape. In two dimensions, however, the zero-phonon processes have a weight zero, but a high concentration of multiphonon processes of very low net energy transfer leads to a low-frequency peak in $P(\omega)$ which remarkably still reflects the natural line shape.

We now briefly discuss the effect of phonon scattering on our result. In three dimensions, the Mössbauer peak remains the Lorentzian,

$$e^{-2W}\frac{\Gamma/\pi}{\omega^2+\Gamma^2} \; ,$$

and one expects merely that the Debye-Waller factor will be modified. In two dimensions, the problem is more complicated, since $P_M(\omega)$ depends strongly on how rapidly the function $f(t)$ diverges. We are not able to say anything definite. A crude estimate of the effect of phonon scattering would be to replace the phonon frequencies ω_k by $\omega_k + i\gamma_k$.

In effect, in Eq. (2.3),

$$\cos\omega t \to \cos\omega t \, e^{-\gamma(\omega)|t|}$$

and, in the denominator of the integrand,

$$\omega^2 \to \omega^2 + \gamma^2(\omega) \; .$$

The fact that $2W$ still diverges[2] indicates that generally $\gamma(\omega)$ must go to zero at least linearly as ω goes to zero. Then, if it goes to zero linearly, e.g.,

$$\gamma(\omega) \to \gamma_0 \omega \; ,$$

the only change in $P_M(\omega)$ is a renormalization of T_Q',

$$T_Q' \to T_Q'(1+\gamma_0^2) \; .$$

If $\gamma(\omega)$ goes to zero faster, there is no change in $P_M(\omega)$. It is important to point out again that the above argument is rough. It may be that phonon interactions will dominate[9] the low-frequency behavior of $P(\omega)$, in which case the Mössbauer effect would be a convenient method of studying phonon interactions.

V. CONCLUDING REMARKS

The 2-d lattice is an instructive case demonstrating that while "long-range ordering" is sufficient, it is by no means necessary for the existence of physical properties which were supposed to be characteristic of the ordered phase. It would be interesting to find real systems[15] for which the 2-d description is accurate. However, the qualitative moral drawn from our simple example is useful also for more complicated systems. Here one should mention the Bragg-like peaks in the structure factor of some lipid systems, recently discussed in terms of a very simplified model by de Gennes.[16] The recently observed[17] Mössbauer effect in a smectic (layered) liquid crystal phase is also an outstanding example.

Finally, we should like to comment on the nature of the order parameter which one should like to define for a 2-d crystal. Even though $\langle X^2\rangle$ "diverges," the motion of an atom in a 2-d lattice is very different from that in a fluid. First, for the lattice $\langle X^2\rangle = O(\ln N)$, while that for a fluid $\langle X^2\rangle = O(N)$. What is more important is the fact that in the harmonic lattice the excursions of the particle, however big, take place around a well-defined average position and nearest-neighbor distances have small fluctuations. In the liquid, however, each particle can cover the whole liquid volume. Once anharmonicity is introduced, diffusion jumps may be allowed among different lattice equilibrium positions. It seems that the crucial characterization of the lattice is that the typical jump diffusion time τ_j be much longer than the longest lattice vibration time $(\omega_D N^{1/2})^{-1} = \tau_0$. As long as $\tau_j \gg \tau_0$ and we observe the lattice on a time scale shorter than τ_j, then after coarse graining over a time comparable to τ_0, it will appear as a static well-defined lattice. This is reminiscent of having superconductivity on a restricted time domain in a thin ("one-dimensional") sample.[5]

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APPENDIX A: SUSCEPTIBILITY $\chi_{\vec{G}}$

We have

$$\chi_{\vec{G}} = N(\langle \rho_{\vec{G}}^2\rangle - \langle \rho_{\vec{G}}\rangle^2) \; , \tag{A1}$$

where

$$N\langle \rho_{\vec{G}}\rangle^2 = N e^{-2W} \tag{A2}$$

and the Debye-Waller factor e^{-2W} is finite in three dimensions but given by [cf. Eq. (2.11)]

$$e^{-2W} = \alpha^{T/T_G} N^{-T/T_G} \tag{A3}$$

in two dimensions. We also have[10]

$$\begin{aligned} N\langle \rho_{\vec{G}}^2\rangle &= N^{-1}\sum_{n,m} \langle e^{i\vec{G}\cdot\vec{r}_n} e^{-i\vec{G}\cdot\vec{r}_m}\rangle \\ &= \sum_n e^{-\langle [\vec{G}\cdot(\vec{x}_n-\vec{x}_0)]^2\rangle/2} \\ &= e^{-2W}\sum_n e^{\langle(\vec{G}\cdot\vec{x}_n)(\vec{G}\cdot\vec{x}_0)\rangle} \; . \end{aligned} \tag{A4}$$

Thus,

$$\chi_{\vec{G}} = e^{-2W}\left[\sum_n e^{\langle(\vec{G}\cdot\vec{X}_n)(\vec{G}\cdot\vec{X}_0)\rangle} - N\right]. \qquad \text{(A5)}$$

We first deal with the 3-d case. In the Debye approximation,

$$\begin{aligned}\ln f_n &\equiv \langle(\vec{G}\cdot\vec{X}_n)(\vec{G}\cdot\vec{X}_0)\rangle \\ &= \frac{TG^2 v^{(3)}}{2\pi^2 mc^3}\int_0^{\omega_D} d\omega \frac{\sin(\omega R_n/c)}{\omega R_n/c} \\ &= \frac{1}{R_n}\frac{TG^2 v^{(3)}}{2\pi^2 mc^2}\int_0^{\omega_D R_n/c} dx \frac{\sin x}{x} \\ &\sim A/R_n \qquad \text{as } R_n \to \infty ,\end{aligned} \qquad \text{(A6)}$$

where $v^{(3)}$ is the volume of the unit cell and

$$A \equiv TG^2 v^{(3)}/2\pi^2 mc^2 . \qquad \text{(A7)}$$

We can then express $\chi_{\vec{G}}$ as

$$\chi_{\vec{G}} = e^{-2W}\sum_n A/R_n + e^{-2W}\sum_n g_n , \qquad \text{(A8)}$$

where

$$g_n \equiv f_n - 1 - A/R_n .$$

The first term diverges as $N^{2/3}$, while the second term can be shown to be of lower order in the limit $N\to\infty$.

In the case of the 2-d Debye model,

$$h_n \equiv \tfrac{1}{2}\langle[\vec{G}\cdot(\vec{X}_n - \vec{X}_0)]^2\rangle = \frac{2T}{T_G}\int_0^{\omega_D} d\omega\left(\frac{1-J_0(\omega R_n/c)}{\omega}\right), \qquad \text{(A9)}$$

where $T_G = 4\pi mc^2/vG^2$. Thus,

$$h_n = \frac{2T}{T_G}\int_0^{\omega_D R_n/c} dx\ \frac{1-J_0(x)}{x} . \qquad \text{(A10)}$$

By integrating by parts we find

$$h_n \to (2T/T_G)\ln(\gamma\omega_D R_n/2c) \quad \text{as } R_n\to\infty , \qquad \text{(A11)}$$

where $\ln\gamma = 0.577\ldots$ is Euler's constant. We replace the sum $\sum_n$ by an integral $2\pi\int dR_n R_n/v$ with an appropriate multiplicative constant and change the variable of integration to $x = R_n/v^{1/2}$, obtaining

$$\begin{aligned}N\langle\rho_{\vec{G}}^2\rangle &= \sum_n e^{-h_n} \\ &= 2\beta^{T/T_G}\int_1^{N^{1/2}} dx\, x^{1-2T/T_G} + \text{term finite} \\ &\qquad\qquad \text{as } N\to\infty \\ &= \beta^{T/T_G}\frac{N^{1-T/T_G}-1}{1-T/T_G} + \text{term finite} \\ &\qquad\qquad \text{as } N\to\infty .\end{aligned} \qquad \text{(A12)}$$

When $1 \gg T/T_G - 1 \gg 1/\ln N$, we have

$$N\langle\rho_{\vec{G}}^2\rangle \approx \text{const}/(T-T_G) . \qquad \text{(A13)}$$

When $T = T_G$, we have

$$N\langle\rho_{\vec{G}}^2\rangle = \ln N. \qquad \text{(A14)}$$

Using (2.8), (2.11), and (A12), we obtain the leading terms of $\chi_{\vec{G}}$ as $N\to\infty$ for temperatures close to or less than T_G,

$$\chi_{\vec{G}} \approx \frac{\beta^{T/T_G}(N^{1-T/T_G}-1)}{1-T/T_G} - \alpha^{T/T_G}N^{1-T/T_G} . \qquad \text{(A15)}$$

We note that χ_G has a maximum which occurs at the temperature

$$T/T_G \approx 1/\ln N,$$

at which point,

$$\chi_{\vec{G}} \approx N/\ln N. \qquad \text{(A16)}$$

APPENDIX B: DETAILED STUDY OF $I(\vec{k},t)$

We have

$$\begin{aligned}I(\vec{k},t) &= N\langle\rho_{\vec{k}}^{*}(t)\rho_{\vec{k}}(0)\rangle \\ &= \frac{1}{N}\sum_{n,m}\langle e^{i\vec{k}\cdot\vec{r}_n(t)}e^{-i\vec{k}\cdot\vec{r}_m(0)}\rangle \\ &= \sum_n e^{i\vec{k}\cdot\vec{R}_n}e^{-h_n(t)} ,\end{aligned} \qquad \text{(B1)}$$

where[10]

$$\begin{aligned}h_n(t) &\equiv -\ln\langle e^{i\vec{k}\cdot\vec{X}_n(t)} - e^{i\vec{k}\cdot\vec{X}_0(0)}\rangle \\ &= \tfrac{1}{2}\langle\{\vec{k}\cdot[\vec{X}_n(t)-\vec{X}_0(0)]\}^2\rangle .\end{aligned} \qquad \text{(B2)}$$

It can easily be shown in the Debye model that

$$h_n(t) = \frac{2T}{T_k}\int_0^{\omega_D} d\omega \frac{1-\cos(\omega t)J_0(\omega R_n/c)}{\omega} , \qquad \text{(B3)}$$

where $T_k \equiv 4\pi mc^2/vk^2$.

We first study $I(\vec{k})$. The behavior of $I(\vec{G})$ has been discussed in Appendix A. We therefore concentrate on the behavior of $I(\vec{k})$ about the Bragg peak $\vec{k} = \vec{G}$. Let

$$\vec{k} = \vec{G} + \vec{\kappa} , \quad \kappa \ll 1/v^{1/2} ;$$

then

$$I(\vec{k}) \sim \int_0^L \frac{dR\,R}{v} e^{-h_n(0)} J_0(\kappa R) . \qquad \text{(B4)}$$

Below T_G, we can replace $h_n(0)$ by its asymptotic behavior for large R_n [cf. (A11)],

$$h_n(0) \to (2T/T_G)\ln(\gamma\omega_D R_n/2c) .$$

Thus, we obtain

$$\frac{I(\vec{k})}{I(\vec{G})} \approx \frac{2(1-T/T_G)}{(\kappa L)^{2(1-T/T_G)}} \int_0^{\kappa L} dx\, x^{1-2T/T_G} J_0(x) . \quad \text{(B5)}$$

Very close to $\vec{k}=\vec{G}$, when $\kappa L \ll 1$, we expand the Bessel function

$$J_0(x) = 1 - \tfrac{1}{4}x^2 + \cdots ,$$

obtaining

$$\frac{I(\vec{k})}{I(\vec{G})} = 1 - \frac{1}{4}\frac{T}{T_G}\left(1-\frac{T}{T_G}\right)\kappa^2 L^2 + \cdots . \quad \text{(B6)}$$

When $\kappa L \gg 1$, the behavior of $I(\vec{k})$ depends upon the ratio T/T_G. When $T/T_G > \frac{1}{4}$, we can replace the upper limit of the integral in (B4) by infinity, obtaining

$$\frac{I(\vec{k})}{I(\vec{G})} \approx \left(1-\frac{T}{T_G}\right)\left(\frac{1}{2}\kappa L\right)^{-2(1-T/T_G)} \frac{\Gamma(1-T/T_G)}{\Gamma(T/T_G)} , \quad \text{(B7)}$$

when $1 > T/T_G > \frac{1}{4}$, and $1 \ll \kappa L \ll N^{1/2}$, where $\Gamma(x)$ is the Γ function.

When $T/T_G < \frac{1}{4}$, the integral diverges as $\kappa L \to \infty$. We can obtain the asymptotic behavior of the integral by using the asymptotic behavior of $J_0(x)$ as $x \to \infty$,[14]

$$\frac{I(\vec{k})}{I(\vec{G})} \approx \pi^{-1/2}\left(\frac{2}{\kappa L}\right)^{3/2} \sin(\kappa L - \tfrac{1}{4}\pi) , \quad \text{(B8)}$$

when $T/T_G < \frac{1}{4}$ and $N^{1/2} \gg \kappa L \gg 1$.

Let us define the quantity

$$I_\Delta(\vec{k}) \equiv \int_{-\Delta}^{\Delta} d\omega\, S(\vec{k},\omega)$$
$$= (\Delta/\pi)\int_{-\infty}^{\infty} dt\, I(\vec{k},t)[\sin(\Delta t)/\Delta t] . \quad \text{(B9)}$$

We can rewrite $h_n(t)$ as

$$h_n(t) = h_n(0) + \frac{2T}{T_k}\int_0^\infty dx \frac{(1-\cos x)J_0(xR_n/ct)}{x}$$
$$- \int_{\omega_D R_n/c}^{\infty} dx \frac{[1-\cos(xct/R_n)]J_0(x)}{x} . \quad \text{(B10)}$$

The important range of R_n values is $R_n \sim L$, while the important range of t values is $t \sim \Delta^{-1}$.

From Ref. 14 we obtain

$$\int_0^\infty dx \frac{(1-\cos x)J_0(xR_n/ct)}{x} = \begin{cases} \cosh^{-1}(ct/R_n), & ct/R_n > 1 \\ 0 , & ct/R_n < 1 . \end{cases} \quad \text{(B11)}$$

Thus, this integral vanishes in the significant regions if

$$c/\Delta L < 1 .$$

When $\omega_D R_n/c \gg 1$, the last integral is on the order of

$$\int_{\omega_D R/c}^{\infty} dx \frac{1-\cos(xct/R_n)}{x} \frac{\cos(x-\frac{1}{4}\pi)}{x^{1/2}}$$
$$< \int_{\omega_D R/c}^{\infty} dx\, x^{-3/2} = 2\left(\frac{c}{\omega_D R}\right)^{1/2} \ll 1 . \quad \text{(B12)}$$

Thus, we expect (we have not proved this rigorously)

$$I(\vec{k},t) \approx I(\vec{k},0) \text{ when } t \ll L/c$$

and thus

$$I_\Delta(\vec{k}) = I(\vec{k}) \text{ when } \Delta \gg c/L .$$

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