Speckle in the Diffraction Patterns of Hendricks-Teller and Icosahedral Glass Models

Anupam Garg and Dov Levine
Institute for Theoretical Physics, University of California, Santa Barbara, Santa Barbara, California 93106
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It is shown that the x-ray diffraction patterns from the Hendricks-Teller model for layered systems and the icosahedral glass models for the icosahedral phases show large fluctuations between nearby scattering wave vectors and from sample to sample, that are quite analogous to laser speckle. The statistics of these fluctuations are studied analytically for the first model and via computer simulations for the second. The observability of these effects is discussed briefly.

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The purpose of this paper is to discuss a phenomenon that is shared by the diffraction patterns of two structural models that aim to describe two very different classes of solids. The first model, proposed by Hendricks and Teller,[1] describes partially ordered layered systems such as graphite, mica, and various clays. It consists of atomic planes with spacings that randomly assume one of two or more values. The second model, called the "icosahedral glass,"[2] arose from an attempt to understand the diffraction patterns from the icosahedral phases discovered by Schechtman *et al.*[3] and Levine and Steinhardt.[4] It consists of identical icosahedral units (representing a cluster of atoms, perhaps), randomly packed together with special attachment rules for near neighbors (face to face, edge to edge, or vertex to vertex) that preserve the orientations of the icosahedra with respect to a fixed set of axes.

The icosahedral glass model is a contender for the geometrical structure of the icosahedral phases for two reasons. First, it produces icosahedrally symmetric diffraction patterns with broad spots, whose location can be made to agree with experiment by adjustment of the size of the icosahedral unit and by correct choice of the attachment rule, and whose widths are intrinsic to the geometry of the model and do not vanish in the infinite-size limit. The latter fact is qualitatively similar to experimental observations even on slowly cooled samples where one might, *a priori*, expect a high degree of structural order.[5] Second, its simplicity makes it appealing from the standpoint of the growth of the phase.[6]

It comes as a surprise that a random aggregate of icosahedra should produce diffraction spots at all—the precise sense in which this is true is discussed below—and Stephens and Goldman argued that this could be understood by analogy with the Hendricks-Teller (H-T) model, which also produces broad spots. Just as the positions of the planes in the H-T model can be described by a directed one-dimensional random walk, the centers of the units in the icosahedral glass model can be described by a branched, self-avoiding random walk in three dimensions, with steps chosen from a set of icosahedral vectors. The projections of these centers onto the direction of the scattering wave vector $\mathbf{Q}$ can be argued, with some plausible assumptions, to form an H-T sequence. The analysis of Hendricks and Teller is then applicable and predicts the positions and widths of the spots.[7,8]

Although the structures produced by both these models are random, one nevertheless expects their physical properties to be well defined in the thermodynamic limit. The main and surprising result of our paper is that this is not so for their diffraction patterns. In other words, the diffraction patterns do not self-average. We shall show that the scattered intensity $I(\mathbf{Q})$ at any wave vector $\mathbf{Q}$ varies from one realization to another in the ensemble, with a variance as large as the mean. Also, the scattered intensities $I(\mathbf{Q})$ and $I(\mathbf{Q}')$ for a given realization are essentially completely uncorrelated for wave vectors $\mathbf{Q}$ and $\mathbf{Q}'$ differing by more than a few times the inverse of the system's linear size. We can prove these results analytically for the H-T model, and we have strong numerical evidence for them in the icosahedral-glass case. The ideal diffraction pattern will therefore show large fluctuations from sample to sample. For a given sample, in addition, one will see large fluctuations between nearby values of $\mathbf{Q}$, lending an extremely noisy, spiky, aspect to the diffraction pattern. This is exactly like the phenomenon of laser speckle,[9,10] except that here the speckle is modulated by the ensemble average, $I_0(\mathbf{Q})$, of the scattered intensity, which is a strongly varying function of $\mathbf{Q}$. Indeed, all previous studies of these models have focused entirely on this average, and it is only in the sense that $I_0(\mathbf{Q})$ has sharp peaks that either model can be said to produce diffraction spots.

Since the H-T model is believed to be a good description of layered systems, the observability of speckle in such materials is subject only to experimental limitations. It is unclear if it can be seen at present, but we see no reason why it should not be feasible with improved technology. Since the icosahedral-glass model is not on quite as firm a footing as the H-T model, the question naturally arises whether observation of speckle in the icosahedral phases could help to distinguish this model from others, such as a defected quasiperiodic structure.[7,11] We shall return to these questions later.

Let us consider the H-T model first. With the assumption of the same form factor for each plane, the normalized scattering intensity (ignoring factors of proportionality) from one realization is given by

$$I(Q)=\frac{1}{N}\sum_i\sum_j e^{iQ(x_i-x_j)}, \tag{1}$$

where the x_i's are the positions of the planes, and N is their number. Since each $\delta_i \equiv x_i - x_{i-1}$ is an independent variable, the ensemble-averaged intensity $\langle I(Q)\rangle \equiv I_0(Q)$ is given in the limit $N\to\infty$ by

$$I_0(Q)=1+\left[\sum_{n=1}^{\infty}\langle e^{iQ\delta}\rangle^n+\text{c.c.}\right], \tag{2}$$

$$=(1-|c|^2)/(1-c)(1-c^*), \tag{3}$$

where $c=\langle e^{iQ\delta}\rangle$.[12]

To study the fluctuations in $I(Q)$, we compute $\langle I^2(Q)\rangle$:

$$\langle I^2(Q)\rangle=\frac{1}{N^2}\sum_{i,j,k,l}\langle e^{iQ(x_i-x_j+x_k-x_l)}\rangle. \tag{4}$$

We can restrict the sum to $x_i<x_k$, $x_j<x_l$, and multiply by 4.[13] It is then merely tedious to verify that the only nonzero contribution as $N\to\infty$ comes from the case where the pair (x_i,x_j) is separated from the pair (x_k,x_l), i.e., $x_i<x_l$ and $x_j<x_k$ in addition to the inequalities above. If we now fix the centers of these pairs and sum over the differences x_i-x_j and x_k-x_l, we get (as $N\to\infty$) two factors of $I_0(Q)$. Summing over the positions of the centers of the pairs gives a factor of $N^2/2$, so that

$$\langle I^2(Q)\rangle=2I_0^2(Q), \tag{5}$$

implying that the variance in $I(Q)$ is as large as the mean, as asserted earlier.[14]

Essentially the same computation yields $\langle I(Q)I(Q')\rangle$. We now associate the pair (x_i,x_j) with Q and (x_k,x_l) with Q', obtaining two independent sums that give $I_0(Q)$ and $I_0(Q')$. There is no longer a factor of 2 arising from the interchange symmetry of the pairs, and so

$$\langle I(Q)I(Q')\rangle\approx I_0(Q)I_0(Q');\quad |Q-Q'|\gtrsim 2\pi/L. \tag{6}$$

The condition that Q and Q' not be too close is clearly necessary since otherwise the pairing $(x_i,x_l),(x_j,x_k)$ would also give a large contribution to the sum. (Here, L is the linear size of the system.)

A simple physical picture for the understanding of the above results is as follows. Let each δ_i be either d_1 or d_2 with probability p_1 and p_2, respectively. Thus the maxima in $I_0(Q)$ occur when $Qd_i\simeq 2\pi m_i$, $i=1,2$, and the m_i are integers. If d_1 and d_2 are incommensurate, however, the phase mismatches, $\phi_i\equiv Qd_i-2\pi m_i$ will never vanish together, implying the absence of Bragg peaks. The choice $\bar\phi=\sum_i p_i\phi_i=0$ can still produce maxima in $I_0(Q)$, nevertheless. The phase (mod 2π) with which a plane scatters performs a random walk, with steps ϕ_1 and ϕ_2, and builds up to a value of π after approximately $n_{\text{coh}}(Q)\sim 1/\langle\phi^2\rangle$ scatterers. We can thus visualize the scattering as taking place from N/N_{coh} coherence domains of size $\xi_{\text{coh}}(Q)\sim N_{\text{coh}}(Q)\bar d$ each, with the scattering within each domain being in phase, and the relative phases between different domains being completely random.[15]

The simple picture presented above is identical to a model for laser speckle,[10] and is easily shown to lead to Eqs. (5) and (6). In fact, it gives the full probability distribution of $I(Q)$:

$$P(I(Q))=I_0^{-1}(Q)\exp[-I(Q)/I_0(Q)]. \tag{7}$$

This can be shown analytically for the H-T model without appeal to the simple speckle picture,[16] and we shall present strong numerical evidence for it in the case of the icosahedral glass.

The analogy between the H-T and the icosahedral glass models suggests that one should see speckle for the latter model too. Since it is difficult to make this argument precise, we have resorted to direct computer simulation. We do this in two dimensions using decagons for simplicity and in order to generate large packings. There is no loss of generality in this and indeed studies performed by us on small packings of icosahedra are consistent with speckle.

We have generated five samples of approximately 250 000 decagons each with the attachment rule that neighboring decagons share an edge. The distance between the centers of such neighbors is taken to be unity, and the number density of our packings is 0.83–0.85, so that the linear size L is approximately 600. Figure 1 shows a radial scan of the diffraction pattern through two of the peaks generated by *one* of these packings. These peaks are located at the same distance $|\mathbf{Q}|$, but are in different directions, and correspond to two symmetry-related spots in a real diffraction pattern. We also show a smoothed average of ten peaks at the same $|\mathbf{Q}|$ obtained from two packings. Note that, as expected, the detailed shapes of the two peaks are very different, and that they are considerably spikier than the average. The width of a spike is about 10^{-2}, consistent with Eq. (6).

Since we would have to generate many more packings than we have in order to test Eq. (5) or (7) directly, we have resorted to the following approach. Suppose one is given n numbers $I_1, I_2, \ldots, I_n$, independently chosen from the distribution (7) for a fixed $\mathbf{Q}$. It can then be shown that the quantities w_i, defined by

$$w_i=\frac{I_i}{(I_1+I_2+\cdots+I_n)/n}, \tag{8}$$

are all identically distributed according to

$$p(w)=[(n-1)/n](1-w/n)^{n-2},\quad w\le n, \tag{9}$$

and that any k of them, $k \leq n-1$, are jointly distributed according to

$$p_k(w_1,\ldots,w_k)=\{(n-1)!/n^k[(n-k-1)!]\}[1-(w_1+\cdots+w_k)/n]^{n-k-1}. \quad (10)$$

If we now examine $I(\mathbf{Q})$ from one sample at five symmetry-related spots with the same $|\mathbf{Q}|$, and hence $I_0(\mathbf{Q})$, the five w_i's constructed according to Eq. (8) should obey Eq. (9) with $n=5$. Since this distribution is independent of the unknown parameter $I_0(\mathbf{Q})$, we can exploit the independence of $I(\mathbf{Q})$ and $I(\mathbf{Q}')$ for large enough $|\mathbf{Q}-\mathbf{Q}'|$ to construct sets of five w's from symmetry-related intensities at several $|\mathbf{Q}|$'s. We have done this by sampling three sets of five symmetry-related peaks. Within each peak we sample 50 $|\mathbf{Q}|$ values at intervals $\Delta|\mathbf{Q}|=0.0402$ to ensure that the intensities are uncorrelated.[17] If $R\equiv n^{-1}\sum_i w_i^2$, then using Eqs. (9) and (10) with $n=5$ and $k=2$, we find that the mean value $\bar{R}$ is $\frac{5}{3}$, and the standard deviation is $5/3\sqrt{14}=0.445$. Using our 50 sets we find a mean value of 1.676, which is well within the standard error, and a standard deviation of 0.417. Indeed, we can check the entire w distribution. Figure 2 shows a histogram of the distribution obtained from all 250 w values for one set of five peaks at the same $|\mathbf{Q}|$ and another from all three sets. The fit by Eq. (9) is quite good, suggesting that Eq. (7) is indeed obeyed.

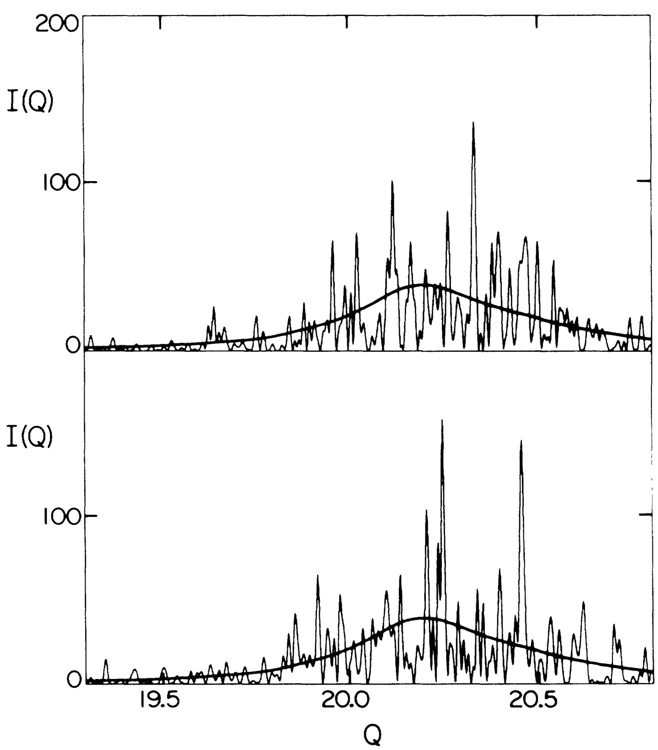

FIG. 1. Two diffraction peaks from one sample of 240000 decagons in two symmetry-related directions at the same value of $|\mathbf{Q}|$. The heavy line is an approximation to $I_0(Q)$ obtained by our smoothing the average intensity of ten peaks at the same $|\mathbf{Q}|$.

Given the ubiquity of speckle in optics, it is natural to ask whether other models[6] for the x-ray diffraction patterns of the icosahedral phases will not also produce speckle. Perhaps the most important of these is the perfect quasicrystal[4] with uniform phason strain.[11,18] By itself this produces only shifts in the peak positions and some additional mechanism for peak broadening must be invoked. A *random* distribution of phason strains accomplishes this; we do not know if the resulting structure produces speckle.

We return finally to the question of observing such speckle experimentally. The foremost requirement is clearly a highly spatially and temporally coherent x-ray source.[19] If the x-ray coherence length is ξ_0, it is necessary to have $\xi_0 \gtrsim L$, and $\xi_0 \gg \xi_{\text{coh}}$, where L is the linear size of the region of the same illuminated by the source. The first requirement is needed to prevent an incoherent addition of several speckle patterns, and the second in order to see the interference between different coherence domains. If we approximate the collective effects of temporal incoherence, absorption, and finite detector resolution by regarding the observed intensity as a convolution of the ideal pattern with a smoothing function of width γ, the observed intensity can be viewed, roughly, as a sum of about $M \simeq (\gamma L/2\pi)^3$ independent random variables, which will reduce the contrast of the speckle, σ_I/I_0, by a factor of $M^{1/2}$, where σ_I^2 is the variance in $I(Q)$. Use of feasible values $\gamma=10^{-3}$ Å^{-1}, $L=10^5$ Å, gives a reduction in contrast by about 60, washing out

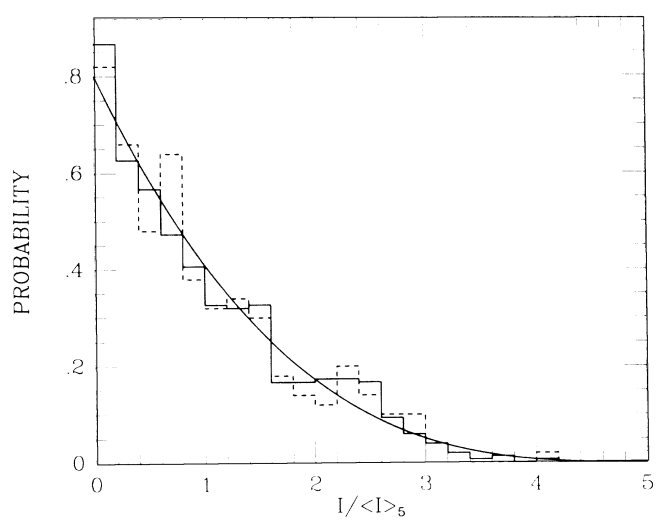

FIG. 2. Histograms of the w distribution [see Eqs. (8) and (9), and following discussion] from one set of peaks (solid line), and from three sets (dashed line). The continuous curve is Eq. (9).

the speckle. We are hopeful that this number can be reduced, thus allowing the observation of these effects.

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[6]V. Elser [in *Proceedings of the Fifteenth International Colloquium on Group Theoretical Methods in Physics,* edited by R. Gilmore and D. H. Feng (World Scientific, Singapore, 1987), Vol. 1] has generated packings akin to those of this model, incorporating considerations of growth.

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[8]The systematics of the peak widths found by the authors of Ref. 5 differ from the theoretical analysis of the simple icosahedral glass presented in Ref. 7. This has prompted variant packings such as those of Ref. 6. It would be interesting to study these for the phenomena we are discussing.

[9]The similarity between the fluctuations we had found and laser speckle was pointed out to us by David DiVincenzo.

[10]For a review of laser speckle, see *Laser Speckle and Related Phenomena,* edited by J. C. Dainty (Springer-Verlag, Berlin, 1984), 2nd Ed., especially the article by J. W. Goodman.

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[12]This derivation assumes that $c(Q)\neq 1$ anywhere. Otherwise $I(Q)$ will possess Bragg peaks with an *unnormalized* strength proportional to N^2 at Q values where $c(Q)=1$.

[13]The cases where $x_i=x_k$, or $x_j=x_l$, or both, can be shown to given zero as $N\rightarrow\infty$.

[14]It is plain that this analysis extends to general, not necessarily directed, random walks, in any number of dimensions, by letting the δ_i's be vectors taking on a finite number of values. Equations (3)-(6) still hold with a suitable redefinition of $c(Q)$.

[15]Note that N_{coh} is Q dependent. By this argument, the maxima of $I_0(Q)$ scale as $N^{-1}(N/N_{\text{coh}})N_{\text{coh}}^2\sim N_{\text{coh}}$, and the widths as ξ_{coh}^{-1}, in complete accord with the Hendricks-Teller analysis.

[16]The argument used to derive Eq. (5) shows that $\langle I^n(Q)\rangle = n!I_0^n(Q)$ implying Eq. (7).

[17]That the intensities are uncorrelated is, strictly speaking, an assumption. We would need many more packings to verify this properly. We believe, however, that a look at Fig. 1 makes this highly plausible, as the mean spacing between maxima is $\simeq 0.02$.

[18]This is inaccurately termed *linear* phason strain in some of the literature.

[19]It is an open question whether speckle can be seen in electron diffraction.