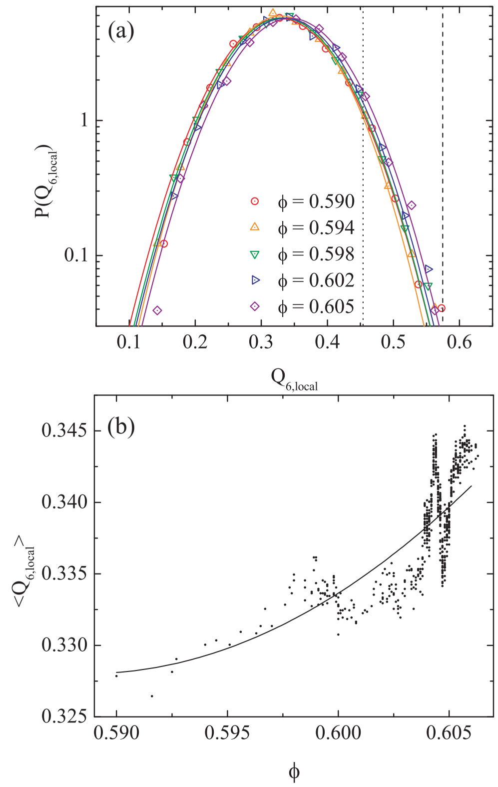

FIG. 3. (a) The probability distribution of Q_6 measured for each particle using Voronoi neighbors are observed to described by Gaussian fits. (b) The mean $Q_{6,local}$ measured as a function of ϕ. The curve is a guide to the eye and shows that Q_6 increase somewhat over the narrow ϕ investigated.

here n is the number of particle sizes in the system. Coarse graining the observed size distribution - shown in Fig. 1(d) - into three sizes $d_1 = 0.98$ mm, $d_2 = 1.02$ mm, $d_3 = 1.07$ mm (using a greater n does not change the results significantly), we calculate the corresponding six distinct $g_{ij}(r)$ terms, which are plotted in the Inset to Fig. 2 for $\phi = 0.59$, and thus the computed $g_{PY}(r)$ for the polydisperse packing according to the Percus-Yevick approximation, which is plotted in Fig. 2. We observe that the amplitude and width of the primary peak and the broad features of the secondary peaks are in good agreement with the Percus-Yevick approximation. This comparison is especially noteworthy because there are no fitting parameters. At higher ϕ, the primary peak and the overall form is still captured by the $g_{PY}(r)$, but details such as the splitting of the second peak which can indicate hexagonal ordering is not captured because Percus-Yevick approximation assumes random angular orientation.

To investigate orientational order in the packing, we use the orientation order metric

$$Q_l \equiv \sum_{i=1}^{N} \left(\frac{4\pi}{l(l+1)} \sum_{m=-l}^{m=l} |\langle Y_{lm}(\Theta(\mathbf{r}), \Phi(\mathbf{r}))\rangle|^2 \right)^{1/2}, \quad (2)$$

where, $l = 6$ to examine hexagonal order, Y_{lm} are the spherical harmonics, $\Theta(\mathbf{r})$ and $\Phi(\mathbf{r})$ are the polar and azimuthal angle, respectively, and $\mathbf{r}$ is the position vector from a particle to its neighbor [15]. We define particle neighbors as those which share a Voronoi cell surface [13]. This removes any ambigity as is introduced when considering only neighbors at contact due to roundoff errors in finding a particle center. In order to compare with packing of elastic particles, we first compute $Q_{6,global}$ by averaging $Y_{lm}(\Theta(\mathbf{r}), \Phi(\mathbf{r}))$ over all the bonds of the packing, and find $Q_{6,global} = 0.27 \pm 0.02$. If particles neighbors are uncorrelated, then Q_6 is small because it goes as square root of the total number of bonds [16]. On the other hand, Q_6 for a FCC crystal with 12 neighbors is 0.5745. But even a slight perturbation due to roundoff errors introduces 2 extra neighbors and the corresponding Q_6 is on average 0.454 [17]. Therefore, the observed distribution shows ordering but the degree of order appears lower compared with simulations of frictionless hard spheres [18], where Q_6 as high as 0.4 was reported for a frictionless hard spheres at comparable ϕ. In those studies which were performed with considerably smaller system size, particle inelasticity was observed to lower Q_6 but not as significantly as in our experiments.

To examine the local orientational order more closely, we plot the observed probability distribution of Q_6 for each particle in Fig. 3(a), and the mean of the distribution $\langle Q_{6,local}\rangle$ in Fig. 3(b). $\langle Q_{6,local}\rangle$ is more sensitive than $Q_{6,global}$ to small crystalline regions within a packing and allows us to avoid the possibility of destructive interference between different crystalline regions [19]. No significant enhancement of distribution is found at the values corresponding to FCC crystal, and the observed Q_6 distribution can be described rather well in fact by Gaussian fits. From these observations we conclude that while there is some local hexagonal order which increases slightly over the ϕ investigated, no significant crystallites occur in this dense regime approaching random close packing.

A complementary method to examine the packing at the particle scale is using the free volume v_f associated with each particle given by subtracting the minimum Voronoi volume corresponding to close packing, $v_c = d^3/\sqrt{2}$ from the Voronoi volume. This statistical quantity has gained prominence because it may be used to define a new measure of entropy based on disorder in packing [6, 20], and may be amenable to thermodynamic interpretation [5]. It has been postulated based on analytical work in 1-dimensional systems, that v_f distribution of random packing of spheres can be described by a Γ distribution [6]:

$$f(v_f) = \frac{\delta\alpha^{(m/\delta^2)}}{\Gamma(m/\delta^2)} v_f^{(m/\delta-1)} e^{-\alpha v_f^{\delta}} \quad (3)$$

with three fitting parameters m, δ and α that control different parts of the distribution and were determined by numerical simulations with frictionless hard spheres [6]. In Fig. 4, we plot v_f normalized by the mean free volume