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Figure S1. Temperature dependent magnetization from SQUID magnetometer given by zero-field cooled (ZFC) and field cooled (FC) curves. The latter measurement is taken with a small applied field of 200 Oe. The response of 20 nm, 100 nm, and 1 um particles are shown in a), b), and c) respectively.

In nanoparticles, a small feature appears around 150 K; previous work attributes this to the presence of unreacted iron precursor residing in the particles; however, we cannot independently confirm that this is the case.¹ Consistent with M-H curves, 100 nm nanoparticles do not appear to have any significant magnetic phase transition across the explored temperature range. The ZFC/FC signature in our microparticle system is representative of the behavior generally seen in superparamagnetic systems. The peak in the ZFC curve at 32 K is known as the blocking temperature, T_B, which refers to the point below which magnetic ordering occurs and above which thermal energy dominates the system's capacity for responding to the applied magnetic field.^{2,3}

Particle size distribution can be quantitatively estimated by observing the distance between the ZFC peak and the point at which the ZFC and FC curves separate, referred to as T_{irr} .² In our microparticle system T_{irr} occurs very near 300 K, indicating a relatively large particle size distribution. Tighter particle distributions will see ZFC and FC separation appear much nearer to T_B .^{2,3}



Figure S2 Size distribution of nano- and microparticles from TEM and SEM, respectively.



Figure S3 UV-visible spectroscopy absorption data for 20 nm, 100 nm, and 1 um particle sizes respectively from which Tauc plots are derived. Sharp features in microparticle absorption data are attributed to chloroform absorption peaks that did were not removed by instrumental background subtraction.



Figure S4 Tauc plot of absorption data for a sample ligated with oleylamine (red) and oleic acid (blue). Observed variation between spectra is due to changes in sample preparation, as different batches of identical material with different concentrations were used. The corresponding Tauc analysis yields consistent band-transition values; calculated indirect band gaps are 0.24 and 0.25 eV, respectively.



Figure S5 Synchrotron XRD pattern of micron-sized particles against reference pattern for cubic FeS_2 from 0 to 30° 20 for the sychrotron wavelength of 0.4146 Å.



Figure S6 Rudimentary model of interfacial area increase with increasing number of grains per particle. Each particle is assumed to be a cube comprised of smaller cubes of equal size. In accordance with the microscopy and Scherrer analyses results presented in the main article, a) single crystalline nanoparticle of the smallest size; b) shows mid-sized particle and c) largest size particle. Cubes are intended as visual aids; the cited dimensions correspond to particle size information described in the text.

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