Equation 12 (i.e. $Pr(\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}|\lambda)$) does not depend on ϵ and σ, the acceptance probability is independent of $Pr(\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}|\lambda)$. Also, as mentioned previously, we use a "weakly informative prior" such that the acceptance probability is independent of $Pr(\epsilon, \sigma|\lambda)$. Furthermore, Q is chosen to be symmetric such that the Q terms in the numerator and denominator of Equation 16 cancel. Therefore, the probability of accepting ϵ_{i+1} and σ_{i+1} is based completely on the likelihood:

$$\alpha = \min\left(1, \frac{L(\epsilon_{i+1}, \sigma_{i+1}|\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda)}{L(\epsilon_i, \sigma_i|\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda)}\right) \tag{17}$$

The likelihood, $L(\epsilon, \sigma, |\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda)$, is calculated from a normal distribution

$$L(\epsilon, \sigma, |\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda) = \prod_{D=\rho_\mathrm{l}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(\rho_\mathrm{l,SM}^\mathrm{sat}(\epsilon,\sigma,\lambda) - \rho_\mathrm{l,D}^\mathrm{sat}\right)^2}{2s_\mathrm{D,SM}^2}\right) \prod_{D=P_\mathrm{v}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(P_\mathrm{v,SM}^\mathrm{sat}(\epsilon,\sigma,\lambda) - P_\mathrm{v,D}^\mathrm{sat}\right)^2}{2s_\mathrm{D,SM}^2}\right) \tag{18}$$

$$L(\epsilon, \sigma, |\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda) = \prod_{D=\rho_\mathrm{l}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(\rho_\mathrm{l,SM}^\mathrm{sat}(\epsilon,\sigma,\lambda) - D\right)^2}{2s_\mathrm{D,SM}^2}\right) \prod_{D=P_\mathrm{v}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(P_\mathrm{v,SM}^\mathrm{sat}(\epsilon,\sigma,\lambda) - D\right)^2}{2s_\mathrm{D,SM}^2}\right) \tag{19}$$

$$L(\epsilon, \sigma, |\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda) = \prod_{D=\rho_\mathrm{l}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(\mathrm{SM}(\epsilon,\sigma,\lambda) - D\right)^2}{2s_\mathrm{D,SM}^2}\right) \prod_{D=P_\mathrm{v}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(\mathrm{SM}(\epsilon,\sigma,\lambda) - D\right)^2}{2s_\mathrm{D,SM}^2}\right) \tag{20}$$

$$L(\epsilon, \sigma, |\rho_\mathrm{l}^\mathrm{sat}, P_\mathrm{v}^\mathrm{sat}, \lambda) = \prod_{D=\rho_\mathrm{l}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(D_\mathrm{SM}(\epsilon,\sigma,\lambda) - D\right)^2}{2s_\mathrm{D,SM}^2}\right) \prod_{D=P_\mathrm{v}^\mathrm{sat}} \frac{1}{\sqrt{2\pi s_\mathrm{D,SM}^2}} \exp\left(\frac{-\left(D_\mathrm{SM}(\epsilon,\sigma,\lambda) - D\right)^2}{2s_\mathrm{D,SM}^2}\right) \tag{21}$$

$$L(\epsilon,\sigma,|\rho_{\mathrm{l}}^{\mathrm{sat}},P_{\mathrm{v}}^{\mathrm{sat}},\lambda)=\prod_{i}^{D=\rho_{\mathrm{l}}^{\mathrm{sat}}}\frac{1}{\sqrt{2\pi s_{\mathrm{D,SM}}^{2}}}\exp\left(\frac{-\left(\rho_{\mathrm{l,SM}}^{\mathrm{sat}}(\epsilon,\sigma,\lambda;T_{i}^{\mathrm{sat}})-\rho_{\mathrm{l,D,i}}^{\mathrm{sat}}\right)^{2}}{2s_{\mathrm{D,SM}}^{2}}\right)$$
$$\prod_{D=P_{\mathrm{v}}^{\mathrm{sat}}}\frac{1}{\sqrt{2\pi s_{\mathrm{D,SM}}^{2}}}\exp\left(\frac{-\left(P_{\mathrm{v,SM}}^{\mathrm{sat}}(\epsilon,\sigma,\lambda)-D\right)^{2}}{2s_{\mathrm{D,SM}}^{2}}\right)\quad(22)$$

$$L(\epsilon,\sigma,|\rho_{\mathrm{l}}^{\mathrm{sat}},P_{\mathrm{v}}^{\mathrm{sat}},\lambda)=\prod\frac{1}{\sqrt{2\pi s_{\mathrm{D,SM}}^{2}}}\exp\left(\frac{-\left(\rho_{\mathrm{l,SM}}^{\mathrm{sat}}(\epsilon,\sigma,\lambda)-\rho_{\mathrm{l,D}}^{\mathrm{sat}}\right)^{2}}{2s_{\mathrm{D,SM}}^{2}}\right)$$
$$\prod\frac{1}{\sqrt{2\pi s_{\mathrm{D,SM}}^{2}}}\exp\left(\frac{-\left(P_{\mathrm{v,SM}}^{\mathrm{sat}}(\epsilon,\sigma,\lambda)-P_{\mathrm{v,D}}^{\mathrm{sat}}\right)^{2}}{2s_{\mathrm{D,SM}}^{2}}\right)\quad(23)$$

$$L(\epsilon,\sigma,|\rho_{\mathrm{l}}^{\mathrm{sat}},P_{\mathrm{v}}^{\mathrm{sat}},\lambda)=\prod_{k}\frac{1}{\sqrt{2\pi s_{\mathrm{D,SM}}^{2}(T_{k}^{\mathrm{sat}})}}\exp\left(\frac{-\left(\rho_{\mathrm{l,SM}}^{\mathrm{sat}}(\epsilon,\sigma,\lambda;T_{k}^{\mathrm{sat}})-\rho_{\mathrm{l,k}}^{\mathrm{sat}}\right)^{2}}{2s_{\mathrm{D,SM}}^{2}(T_{k}^{\mathrm{sat}})}\right)$$
$$\prod_{k}\frac{1}{\sqrt{2\pi s_{\mathrm{D,SM}}^{2}(T_{k}^{\mathrm{sat}})}}\exp\left(\frac{-\left(P_{\mathrm{v,SM}}^{\mathrm{sat}}(\epsilon,\sigma,\lambda;T_{k}^{\mathrm{sat}})-P_{\mathrm{v,k}}^{\mathrm{sat}}\right)^{2}}{2s_{\mathrm{D,SM}}^{2}(T_{k}^{\mathrm{sat}})}\right)\quad(24)$$

where the first and second products are over the experimental $\rho_{\mathrm{l}}^{\mathrm{sat}}$ and $P_{\mathrm{v}}^{\mathrm{sat}}$ data, respectively, T_{k}^{sat} is the saturation temperature that corresponds the k^{th} data point, "SM" refers to the surrogate model (see Section IV B) used to estimate $\rho_{\mathrm{l}}^{\mathrm{sat}}$ or $P_{\mathrm{v}}^{\mathrm{sat}}$ for a given ϵ,σ, λ, and T^{sat}, and $s_{\mathrm{D,SM}}^{2}$ is the combined variance of the experimental data and the surrogate model. The variances are independent, meaning that the combined variance is the sum of the experimental and surrogate model variances, i.e. $s_{\mathrm{D,SM}}^{2}=s_{\mathrm{D}}^{2}+s_{\mathrm{SM}}^{2}$.[48]

B. Surrogate Model

A typical Markov Chain requires $O(10^4$ to $10^5)$ Monte Carlo steps, where the likelihood function must be evaluated at each step. Since $L(\theta|D,M)$ depends on the force field parameters (ϵ,σ, and λ), an MCMC approach is computationally infeasible if computing $L(\theta|D,M)$ requires performing direct molecular simulations for every proposed parameter set. Furthermore, propagation of uncertainty with robust posterior prediction may require $O(10^2$ to $10^3)$ θ_{MCMC} parameter sets for adequate representations of