Predicting the Viscosity of a Supercooled Liquid XI LIN, Boston University — We present an atomistic description of the viscosity of a supercooled liquid capturing the highly non-Arrhenius temperature variation for which no previous calculation has been given. A temperature dependent activation energy for structural relaxation is derived by mapping the potential energy surface and extracting saddle-point configurations and associated atomic coordinates. This essential information is combined with the temperature variation of an effective local energy minimum (inherent structure) to describe shear relaxation by thermal activation. For a binary Lennard-Jones model the calculated viscosity shows a characteristic crossover from strong (Arrhenius) to fragile (highly non-Arrhenius) behavior upon appreciable undercooling, followed by a second crossover from fragile back to strong behavior on approaching the glass transition temperature, both features we believe to be generic. Analysis of atomic displacements associated with barrier crossing in the fragile regime suggests a scenario of correlated motions along a chain of particles as the underlying mechanism for slow viscous relaxation in glassy states.

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Date submitted: 05 Dec 2007