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Gaussian Molecular Dynamics in Imaginary and Real Time
IONUT GEORGESCU, VLADIMIR MANDELSHTAM, University of California, Irvine — The variational Gaussian wavepacket (VGW) method can be used to estimate the equilibrium density matrix by propagating Gaussian wavepackets in imaginary time [1,2]. It has proven to be practically accurate and computationally less expensive than the path integral methods. We compare the VGW method to the Feynman-Kleinert approximation (FKA), which has comparable computational cost. Although both methods are variational, they utilize different variational principles: In FKA the partition function is optimized, while in VGW it is the imaginary-time-dependent wave packet. We show that the VGW method is more accurate for a wide variety of systems. The differences are particularly important when thermodynamic properties, such as heat capacity, are of main interest. Moreover, unlike the case of FKA, in the VGW method the imaginary frequencies do not arise. In the spirit of the Centroid Molecular Dynamics the VGW method has also been extended to simulate the real-time dynamics, e.g., it can be used to estimate the Kubo-transformed quantum time correlation functions. The latter are exact in the high-temperature and harmonic limits.
[1] P. Frantsuzov and V.A. Mandelshtam, J. Chem. Phys **121**, 9247 (2004)
[2] C. Predescu, P. Frantsuzov and V.A. Mandelshtam J. Chem. Phys **122**, 154305 (2005)

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