

Comment on "Local frequency analysis and the structure of classical phase space of the LiNC/LiCN molecular system" [J. Chem. Phys. **108**, 63 (1998)]

C. C. Martens

Department of Chemistry, University of California, Irvine, California 92697

M. J. Davis

Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439

G. S. Ezra

Department of Chemistry, Cornell University, Ithaca, New York 14853

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Losada *et al.*¹ have recently applied the method of local frequency analysis (LFA),^{2,3} as implemented by Laskar,⁴ to study the classical phase space structure and dynamics in a two degree of freedom model for the floppy molecule LiCN/LiNC. The purpose of the present comment is twofold.

First, we note that the fast Fourier transform (FFT)-based approach to LFA as implemented in Refs. 2 and 3 is not a "standard" FFT method, as asserted in Ref. 1. In fact, the method involves fitting peak line shapes to a known form to obtain significantly higher frequency resolution (a factor of ~ 400) than standard FFT methods. Details are given in Refs. 3, 5, and 6.

Second, contrary to the assertion by Losada *et al.* that "it is not clear *a priori* if the LFA method would render useful information on the dynamics of this type of system" (Ref. 1), our 11-year-old work on the OCS molecule already

made it quite clear that applying LFA to strongly coupled molecular systems (*2d* and *3d*) indeed renders much useful information;^{2,3} in addition to quantitative analysis of quasi-periodic motion, resonant segments of irregular trajectories were identified to high accuracy. Moreover, crossing of irrational frequency ratios (cantori) was clearly seen.³

¹J. C. Losada, J. M. Estebarez, R. M. Benito, and F. Borondo, *J. Chem. Phys.* **108**, 63 (1998).

²C. C. Martens, M. J. Davis, and G. S. Ezra, *Chem. Phys. Lett.* **142**, 519 (1987).

³C. C. Martens, Ph.D. thesis, Cornell University, 1987.

⁴J. Laskar, *Icarus* **88**, 266 (1990); J. Laskar, C. Froeschlé, and A. Celletti, *Physica D* **56**, 253 (1992); J. Laskar, *ibid.* **67**, 257 (1993); H. S. Dumas and J. Laskar, *Phys. Rev. Lett.* **70**, 2975 (1993).

⁵C. C. Martens and G. S. Ezra, *J. Chem. Phys.* **83**, 2990 (1985).

⁶C. C. Martens and G. S. Ezra, *J. Chem. Phys.* **86**, 279 (1987).