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Controlling vibrational excitations in C_{60} by laser pulse durations GUOPING ZHANG, Department of Physics, Indiana State University, Terre Haute, IN 47809, THOMAS F. GEORGE, Office of the Chancellor — Two similar off-resonant ultrafast laser experiments [1-3] in C_{60} have reported two different vibrational modes that dominate the relaxation process: one predicts the ag modes while the other the hg modes. A systematic simulation presented here reveals that this experimental discrepancy results from the laser pulse duration. The numerical results show that since each mode ν has a distinct optimal duration τ_o^ν, the ag modes are strongly suppressed for durations longer than 40 fs, while the hg modes start to grow. For the off-resonant and low-intensity excitations, the period Ω_ν^o of the dominant mode and τ_o^ν satisfy the relation $\Omega_\nu^o/\tau_o^\nu \approx 3.4$. By carefully scanning the laser frequencies and pulse durations, a comprehensive excitation diagram is constructed, which can be used to guide experiments to selectively excite the ag and hg modes in cm by an ultrafast laser [4,5]. Its potential impact is also discussed. [1] S. Dexheimer *et al.*, *Ultrafast Phenomena VIII*, edited by J. L. Martis *et al.*, *Springer Series in Chemical Physics* **55**, 81 (1993). [2] V. R. Bhardwaj *et al.*, *Phys. Rev. Lett.* **91**, 203004 (2003). [3] H. Hohmann *et al.*, *Phys. Rev. Lett.* **73**, 1919 (1994). [4] G. P. Zhang and T. F. George, *Phys. Rev. Lett.* **93**, 147401 (2004). [5] G. P. Zhang, *Phys. Rev. Lett.* **91**. 176801 (2003).

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