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Maximizing the response of a specific vibrational mode to a laser pulse CHARLES SHANNON, ROLAND ALLEN, Texas A&M University — In previous publications [1,2] our group predicted the following for molecules and materials responding to femtosecond-scale optical laser pulses: The maximum relative excitation of a Raman-active vibrational mode with period T will be attained when the pulse has a full-width-at-half-maximum duration $\tau \approx 0.42\,T$. The analytical model used for this prediction involved averaging over the oscillations of the field within the pulse, and the absolute (rather than relative) response is maximized as $\tau \to 0$. Here we generalize the model to include the oscillations of the field, and we find that the absolute maximum is shifted to a nonzero value of the duration which depends on the other parameters of the laser pulse, as well as the period of the vibrational mode. This result is obtained analytically (using Mathematica) and is confirmed by numerical calculations.

[1] Xiang Zhou, Zhibin Lin, Chenwei Jiang, Meng Gao, and Roland E. Allen, Physical Review B 82, 075433 (2010); arXiv:1001.1016 [cond-mat].
[2] Chenwei Jiang, Xiang Zhou, Ruihua Xie, Fuli Li, and Roland E. Allen, Chemical Physics Letters 515, 137 (2011).

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