Theoretical aspects of femtosecond double-pump single-molecule spectroscopy. I. Weak-field regime

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Electronic Supplementary Information

CONTENT

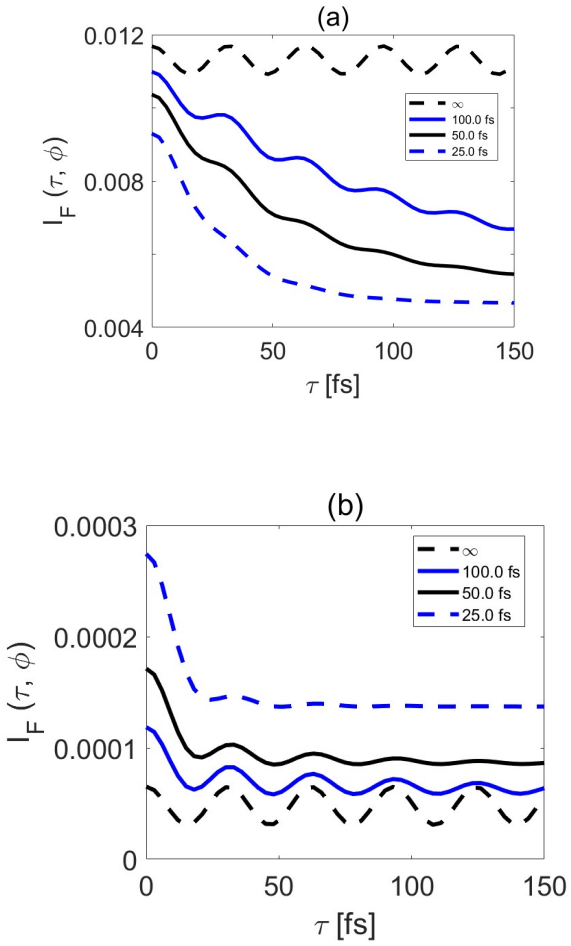

Figure 1: SM signal $I_F(\tau, 0)$ vs. τ for different dephasing times indicated in the legend. The pump pulses are weak ($E = 0.0025$ eV) and short ($\tau_p = 10$ fs), their relative phase is set to zero ($\phi = 0$). The panels corresponding to different shifts of the potential energy surface functions. (a): $\Delta = 1$. (b): $\Delta = 4$.

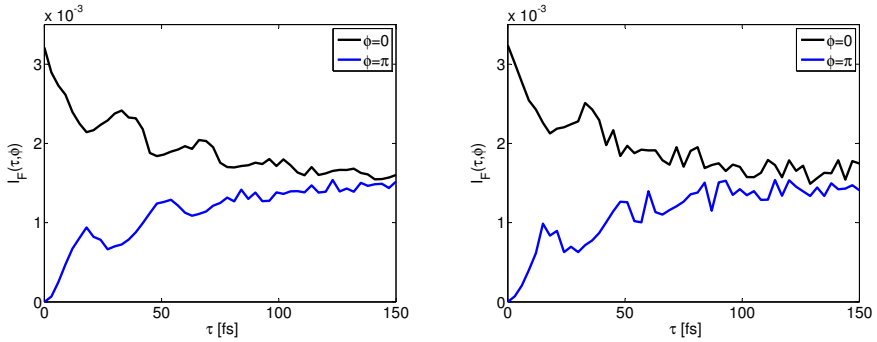

Figure 2: SM signal $I_F(\tau, 0)$ (black line) and $I_F(\tau, \pi)$ (blue line) for different amplitudes of the vibrational frequency modulation: $\delta_{\Omega_2} = 0.014$ eV (left panel) and 0.028 eV (right panel). The remaining parameters have the mean values as specified for Fig. 5(a) of paper I.

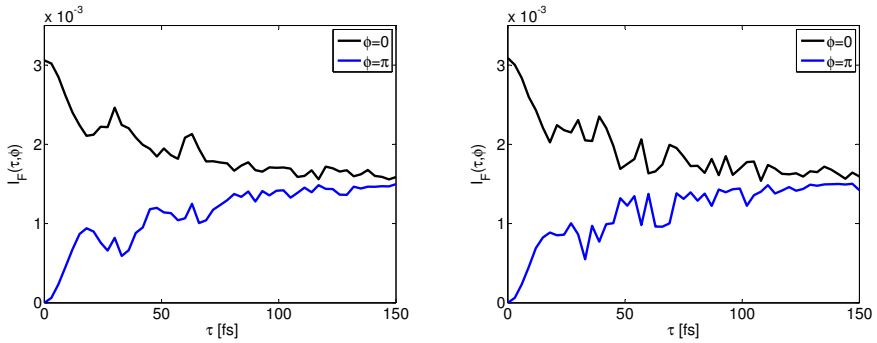

Figure 3: SM signal $I_F(\tau, 0)$ (black line) and $I_F(\tau, \pi)$ (blue line) for different amplitudes of the modulation of the electronic dephasing time, $\delta_{\gamma^{-1}} = 30$ fs (left panel) and 50 fs (right panel). The remaining parameters have the mean values as specified for Fig. 5(a) of paper I.

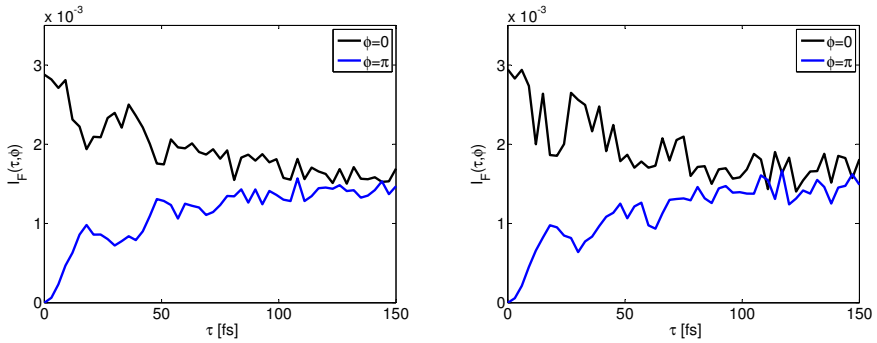

Figure 4: SM signal $I_F(\tau, 0)$ (black line) and $I_F(\tau, \pi)$ (blue line) for different modulations of the shift of the potential energy function: $\delta_\Delta = 0.12$ (left panel) and 0.2 (right panel). The remaining parameters have the mean values as specified for Fig. 5(a) of paper I.

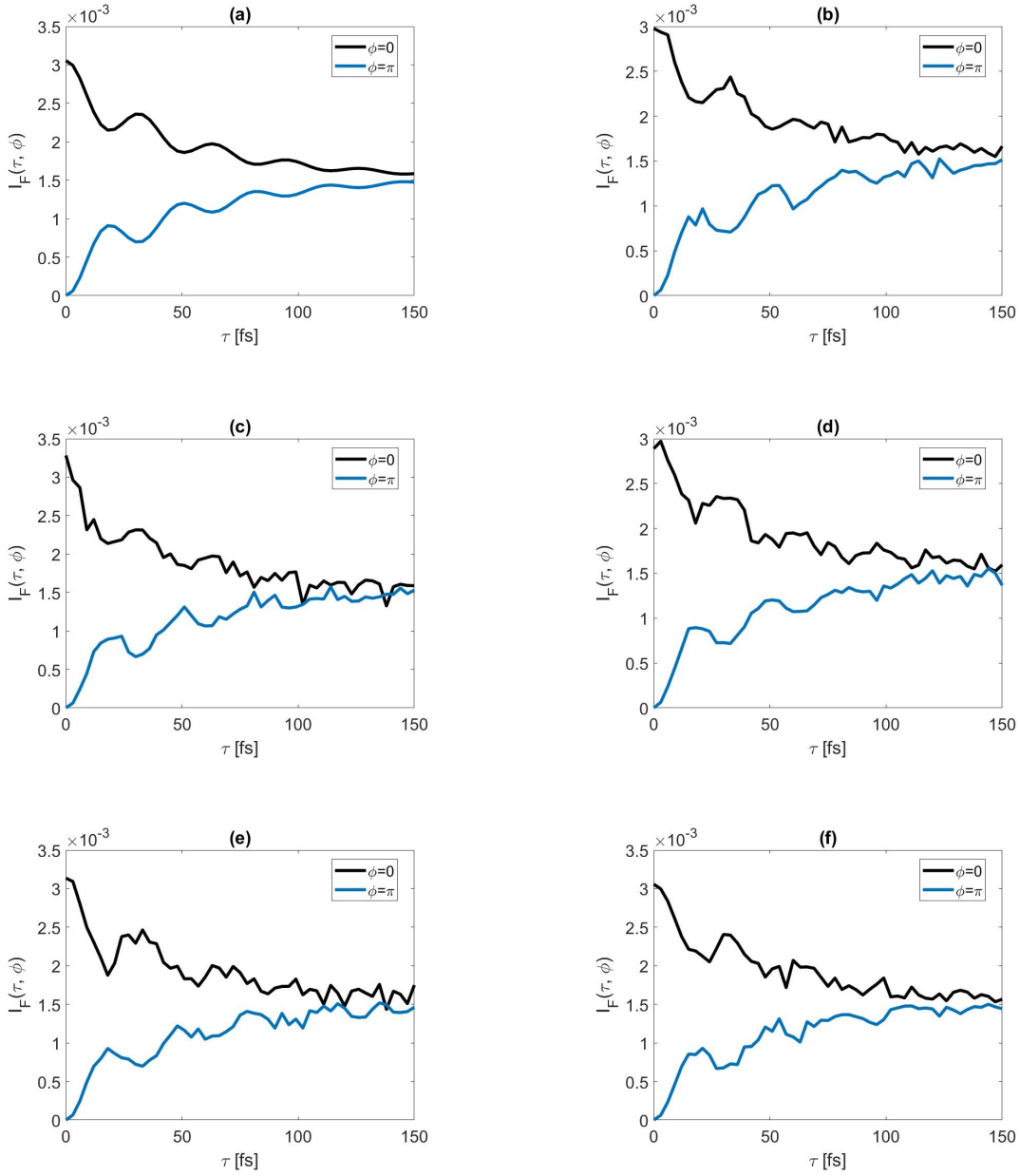

Figure 5: Panel (a) depicts the SM signal $I_F(\tau, 0)$ (black line) and $I_F(\tau, \pi)$ (blue line) evaluated for the following mean values of the parameters: $\bar{\Omega} = 0.13$ eV, $\bar{\omega}_{det} = 0$, $\bar{\Delta} = 2$, $\overline{\hat{\boldsymbol{e}}\hat{\boldsymbol{\mu}}} = 1$, and $\bar{\gamma}^{-1} = 50$ fs. Panels (b) through (f) show the effect of a Gaussian modulation of the vibrational frequency, $\delta_\Omega = 0.0035$ eV (b); electronic energy, $\delta_\epsilon = 0.005$ eV (c); displacement of the potential energy function, $\delta_\Delta = 0.02$, (d); molecular orientation, $\hat{\boldsymbol{e}}\hat{\boldsymbol{\mu}} = \cos(\pi/4 - \varphi)/\cos(\pi/4)$, $\delta_\varphi = 0.05$ (e); electronic dephasing, $\delta_{\gamma^{-1}} = 10$ fs (f). The signals are evaluated with a discretization step of 3 fs in τ. To have the same variance $\langle (A - \bar{A})^2 \rangle$ of the parameters for the uniform and Gaussian distributions, the amplitudes δ_A for the present figure are twice smaller than those for Fig. 5 of paper I.

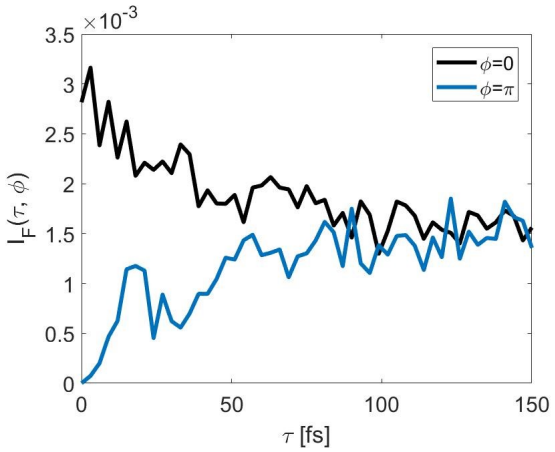

Figure 6: SM signals $I_F(\tau, 0)$ (black line) and $I_F(\tau, \pi)$ (blue line) evaluated with the account of simultaneous Gaussian modulations of the parameters $A = \Omega, \epsilon, \Delta, \varphi, \gamma$. The numerical values of δ_A are indicated in caption for Fig. 5.