Gain-dispersion coupling induced by transient light shifts in an atomic medium

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A weak resonant femtosecond pulse propagates through an optically thick atomic medium whose properties have been modified by a strong nonresonant ultrashort pulse. We study the energy redistribution between the spectral components of the weak resonant pulse using both experimental and theoretical methods. The transmitted spectrum exhibits an asymmetric feature that reveals an important gain-dispersion coupling. For light shifts that perturb slightly the propagation of the weak resonant pulse, this asymmetry maps out the asymptotic part of the linear susceptibility.

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I. INTRODUCTION

When an optical field propagates in an optically thick medium, it experiences both dispersion and absorption (or gain). The spectral behavior of the transmitted field is determined by the spectral response $\mathrm{Re}(\omega,\omega')$ that links the radiated field $E_{\mathrm{rad}}(\omega)$ at ω and the excitation field $E_{\mathrm{exc}}(\omega')$ at ω', given by $E_{\mathrm{rad}}(\omega)=\int_{-\infty}^{+\infty}\mathrm{Re}(\omega,\omega')E_{\mathrm{exc}}(\omega')d\omega'$. When the medium properties are time-invariant (stationary), the response function is diagonal $[\mathrm{Re}(\omega,\omega')=\mathrm{Re}_0(\omega)\delta(\omega-\omega')]$. Then, the real and imaginary parts induce a phase shift (dispersion) and an amplitude modification (absorption or gain) to the transmitted field, respectively. These two quantities are connected by causality through the Kramers-Kronig relations [1] that play a central role in the interpretation of experiments each time the causality is called into question, as it is the case in superluminal propagation experiments [2]. When the medium is not stationary, the response function is no longer diagonal, and the modifications of the amplitude and the phase of each frequency component involve the contribution of all the other parts of the pulse spectrum. Generally, the interplay between the different components leads to complex coupling with the appearance of spectral features that can hardly be interpreted. In this paper, we consider a two-level system whose optical properties are modified by the application of a *strong nonresonant ultrashort pulse* that induces lightshifts and we focus on the spectral reshaping effects experienced by *resonant propagating ultrashort pulse*. Little attention has been paid to such a situation, [3–5] in strong contrast with extensive studies performed in the monochromatic limit [6]. We show that for small lightshifts two important features appear. First, although the dynamics is intrinsically transient, the system exhibits an effective spectral response that is asymptotically diagonal. Secondly, the off-resonant (asymptotic) part of the differential spectrum exhibits dispersionlike features characterized by a decreasing behavior similar to that of the linear susceptibility obtained in the stationary situation (when the driving pulse is not applied). In consequence, the asymptotic part of the linear susceptibility is then directly imprinted on the transmitted spectrum. The situation where the excitation sequence consists of ultrashort pulses has been studied by the authors theoretically for lightshifts such that new frequency bands are generated allowing the shaping of the resonant pulse at a time scale smaller than its initial duration [4]. In this paper, we consider the case of small lightshifts that modify only slightly the propagation effect. No new frequency components are created and the interplay between the gain and dispersion can be clearly established. The appearance of dispersion-like features in the transmitted spectrum is often reported in the monochromatic regime [7] and is a consequence of the competition between quantum paths involving amplification and absorption in the dressed-state picture. Here, only a single quantum path involving absorption is involved in the process and the observed structure thus has a different physical origin. It is shown to be *a direct consequence of the transient character of the lightshifts*. The results obtained point out the crucial role of the light-shift and pave the way for further interpretations of experiments where the spectral reshaping behavior can play an important role. This is the case of spectral modifications that experience a strong resonant pulse during propagation and that are still not completely interpreted [8,9].

II. THEORETICAL MODEL

We consider a sample of two-level systems $(|a\rangle,|b\rangle)$ with energies 0 and $\hbar\omega_0$, respectively. A weak resonant ultrashort pulse (referred as the propagating pulse) propagates through the medium. Its electric field is expressed as $E_p=[\varepsilon_{0p}f_p(z,t)e^{-i(\omega_o t-k_p z)}+\mathrm{c.c.}]/2$ where $f_p(z,t)$ is the envelope, with $f_p(0,t)=(1/\sqrt{\pi})e^{-(t/\tau_p)^2}$ and τ_p is the temporal width of the pulse at the entrance of the sample. When the optical medium is thick, strong reshaping effects distort the temporal profile of the pulse [10]. In the spectral domain, the frequency components experience only a phase shift due to dispersion and the spectrum amplitude is almost unaffected by absorption. Indeed, the pulse is short so the spectrum width τ_p^{-1} is larger than the absorption line given by the Doppler width (Δ_{dop}), so most of the initial energy is transmitted. In the following, we study the changes induced in the spectrum of the propagating pulse by the action of a strong pulse (referred as the driving pulse) with electric field $E_d=[\varepsilon_{0d}f_d(z,t)e^{-i(\omega_d t-k_d z)}+\mathrm{c.c.}]/2$ where $f_d(0,t)=(1/\sqrt{\pi})e^{-(t/\tau_d)^2}$ [Fig. 1(a)]. We assume that the detuning

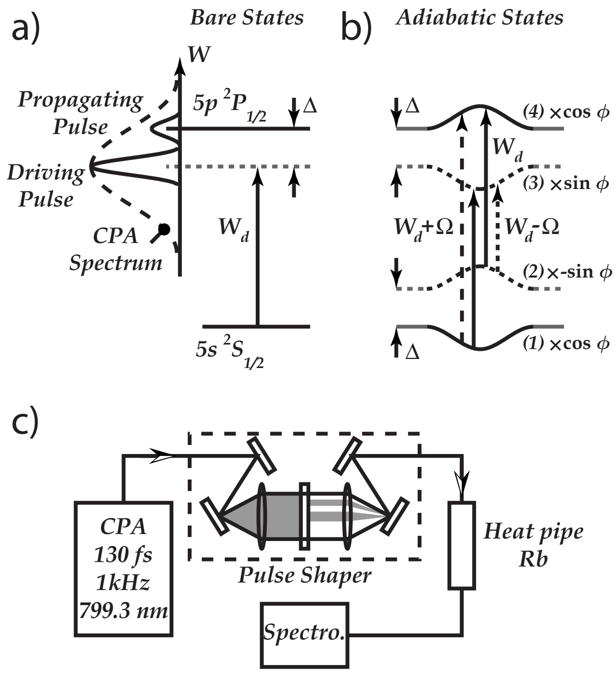

FIG. 1. (a) Excitation scheme of the two-level system (transition $5s^2S_{1/2}\rightarrow 5p^2P_{1/2}$ in the rubidium atom) by the bichromatic field (issue experimentally from a single femtosecond pulse). The strong nonresonant ultrashort pulse modifies the optical response of the system that at the same time probes the weak resonant ultrashort pulse. (b) Adiabatic representation of the two-level system. In our case, the propagating pulse preferentially connects levels (1) and (4). (c) Experimental setup.

$\omega_d-\omega_0$ and the strength of the driving pulse are high enough to prevent any alteration of the strong field caused by its interaction with the atomic medium [i.e., $f_d(z,t)=f_d(0,t)$]. We introduce dimensionless time, space, and frequency variables as $T=(t-z/c)/\tau_p$, $Z=z/L$, $W_d=\omega_d\tau_p$, $W_o=\omega_o\tau_p$, and $\Delta=(W_0-W_d)\tau_{dp}$ (L is the length of the sample, $\tau_{dp}=\tau_d/\tau_p$). We also define by $\theta_j=\mu_{ab}\varepsilon_{0j}\tau_j/\hbar$, $j=p,d$ the pulse areas that characterize the strength of the pulses; $\Omega=\sqrt{\theta_d^2f_d^2+\Delta^2}$ the dimensionless generalized Rabi frequency and μ_{ab} the dipole moment. We assume that the rotating wave approximation is valid in our situation and the durations of the two pulses are comparable.

The interaction process between the atomic system and the two-pulse sequence can be clearly exhibited in the adiabatic representation associated with the driving pulse. The adiabatic states are defined by the following relations:

$$|-\rangle(T)=\cos\phi(T)|a\rangle+\sin\phi(T)e^{-iW_dT}|b\rangle,$$

$$|+\rangle(T)=-\sin\phi(T)|a\rangle+\cos\phi(T)e^{-iW_dT}|b\rangle, \qquad (1)$$

with $\tan(2\phi)=(\theta_d/\Delta)f_d(0,T)$. The energy of the states $|\pm\rangle$ are $\frac{1}{2}(\Delta\pm\Omega)$, respectively. The coupling between these states is given by $\partial_T\phi$ and can be neglected for large detuning ($\Delta\gg 1$) as will be assumed throughout the rest of the paper. The adiabatic levels undergo a transient light-shift during the action of the driving pulse. If all the population initially is in the ground state $|a\rangle$, no transfer of population to state $|+\rangle$ is possible. In the adiabatic basis, the wave function reads as $|\psi\rangle(Z,T)=\alpha_-(Z,T)|-\rangle(T)+\alpha_+(Z,T)|+\rangle(T)$, and we can approximate the zero order amplitudes $\alpha_\pm^{(0)}$ (with respect to the propagating field) by their expression in the adiabatic limit:

$$\alpha_-^{(0)}(Z,T')\simeq e^{-i\int_{-\infty}^{T}[(\Delta-\Omega)/2]d(T'/\tau_{dp})}, \qquad (2a)$$

$$\alpha_+^{(0)}(Z,T')\simeq 0. \qquad (2b)$$

As the weak pulse is applied, transitions between adiabatic states occur [Fig. 1(b)] whereas the shape of the pulse evolves according to the equation of propagation [11]:

$$\frac{\partial}{\partial Z}f_p=ie^{i\Delta T/\tau_{dp}}\frac{e_{\text{disp}}}{\theta_p}\rho_p. \qquad (3)$$

Here, we have neglected the diffraction effects, the Doppler effect and used the slowly varying envelope approximation. The term on the right-hand side of Eq. (3) represents the envelope of the radiated field (per unit of length) and is proportional to the coherence $\rho_p=\langle\psi|a\rangle\langle b|\psi\rangle e^{iW_dT}$. The dimensionless coefficient $e_{\text{disp}}=n\mu_{ab}^2\omega_dL\tau_p/c\varepsilon_0\hbar$ characterizes the severity of propagation effects on the $|a\rangle-|b\rangle$ transition and depends on the density of atoms n and the oscillatory strength through the square of the dipole moment μ_{ab}^2. The expression of the coherence ρ_p in Eq. (3) can be derived using the adiabatic representation [Fig. 1(b)]. Using relations (2–a, 2–b) four quantum paths contribute to the coherence ρ_p corresponding respectively to absorption of the propagating pulse from level 1 to levels 3 and 4, and to emission from level 3 to levels 1 and 2. The associated oscillation frequencies are $W_d(1\leftrightarrow 3)$, $W_d-\Omega(3\rightarrow 2)$ and $W_d+\Omega(1\rightarrow 4)$. Considerable simplification can be made using the fact that the propagating pulse whose frequency matches the atomic frequency of the two-level system is no longer resonant with the system when the driving pulse is applied. Because of the large detuning ($\Delta\gg 1$), the dominant contribution originates from path $1\rightarrow 4$ for which the transition frequency is the closest to the propagating pulse frequency. Within these simplifications, the effect of the driving field reduces to only a transient lightshift of levels 1 and 4. The coherence ρ_p is then reduced to $\rho_p\simeq\alpha_-^{(0)^*}\alpha_+^{(1)}$, where $\alpha_+^{(1)}$ is the amplitude α_+ to the first order with respect to the weak propagating field. The spectral behavior of the radiated field can be understood by performing a Fourier transform of relation (3). From Appendix A, it follows that

$$\frac{\partial\hat{f}_p}{\partial Z}(Z,W)=\int_{-\infty}^{+\infty}\hat{f}_p(Z,W')\text{Re}(W,W')dW', \qquad (4)$$

with $\text{Re}(W,W')=ik(2\pi)^{-2}[\chi(W)\hat{L}(W-W')]\otimes\hat{L}(-W)$ the spectral response of the system and $k=k_p=L/2$. Here, $\hat{f}$ denotes the Fourier transform of f, $\otimes$ is the convolution operation, $\chi(W)=i(e_{\text{disp}}/2k)\hat{h}(W)$ is the linear susceptibility of the system with $\hat{h}(W)=\lim_{\Gamma\rightarrow 0}[1/(\Gamma-iW)]=\pi\delta(W)+i\,\text{P}(1/W)$ and δ and P denote the Dirac function and the Cauchy principal value, respectively [$h(T)$ is the Heaviside function].

$L(T)=e^{i\int_{-T}^{T}[I_s(T')/2]d(T'/\tau_{dp})}$ [with $I_s(T)=\Omega(T)-\Delta$] characterizes the action of lightshifts with $L(+\infty)[L^*(+\infty)]$ representing the adiabatic phase associated with state $|-\rangle[|+\rangle]$, respectively. Note that in our model we have neglected both the relaxations and the Doppler effects, so the susceptibility that appears here does not exhibit the exact shape of the linewidth very close to the resonance. When no driving pulse is applied, $L(T)=1$ and $\mathrm{Re}(W,W')=ik\chi(W)\delta(W-W')$. No coupling between the frequency components occurs. Only the resonance frequency $W=0$ is absorbed, whereas the other frequencies experience a phase shift that scales as $1/W$ (dispersion). Moreover, for $W\neq 0$, the transmitted amplitude spectrum (at zero order with respect to the weak field) is $\hat{f}_p^{(0)}(1,W)=\hat{f}_p^{(0)}(0,W)e^{-i(e_{\mathrm{disp}}/2W)}$. When $e_{\mathrm{disp}}\geq 1$, the dispersion effects affect the whole part of the propagating pulse spectrum. Since the Doppler effect and the relaxations are neglected in our model, the absorption line reduces to a Dirac function centred at the resonance frequency $W=0$. Taking the Doppler effect into account, we remove the singularity and we obtain $\hat{f}_p^{(0)}(1,0)=\hat{f}_p^{(0)}(0,0)e^{-e_{\mathrm{disp}}/2\Delta_{dop}\tau_p}$ [12]. The transmitted amplitude for $W=0$ is very small as long as $e_{\mathrm{disp}}\gg\Delta_{\mathrm{dop}}\tau_p$. This can be obtained even if $e_{\mathrm{disp}}\ll 1$ for which the dispersion effects negligible.

When the driving pulse is applied, strong coupling appears between the spectral frequencies of the propagating pulse and it is consequently no longer possible for arbitrary light-shifts to define a gain or dispersion coefficient. Energy redistribution of the initial pulse components causes new frequencies and side bands to appear [4]. In the general case, it is not possible to give a simple physical picture for all the spectral features that appear from the solution of Eq. (4). We restrict the study in the following to small lightshifts for which there is no significant creation of new spectral components. We will show the experimental result for the transmitted intensity of the propagating pulse that exhibits an asymmetrical distortion. We will provide an interpretation of this structure using the formalism developed in this section.

III. EXPERIMENT AND INTERPRETATION

The experiment was performed in atomic rubidium, on the $5s^2S_{1/2}\rightarrow 5p^2P_{1/2}$ transition (resonance at 794.76 nm). A regenerative amplifier pumped by a titanium sapphire laser provides optical pulses with about 130 fs duration (FWHM of the intensity) and a spectrum bandwidth (FWHM) of 8.5 nm. The laser beam is sent successively into the pulse shaper and a 12 cm long heat pipe containing an atomic vapour of rubidium [Fig. 1(c)]. The pulse shaping device is a $4f$ zero-dispersion line setup composed of one pair each of reflective gratings and cylindrical mirrors. Its active elements are constituted by a combination of two 640-pixels liquid-crystal spatial-light modulators (LC-SLM Ienoptik) located in the common focal plane of both mirrors of the $4f$ line [13]. Each pixel is 100 μm width. The LC-SLM has a high spectral resolution in both phase and amplitude. Two Gaussian spectral bands centred at resonance and at 799.3 nm are cut off from the input spectrum and represent the propagating and driving pulse, respectively (with $\Delta\simeq 10$). The high spatial dispersion of the pulse shaper (0.06 nm/pixel) allows us to achieve bandwidths as narrow as 1.8 and 1.2 nm, respectively, corresponding to pulse durations of $\tau_p=545$ fs and $\tau_d=815$ fs (so $\tau_{dp}\simeq 1.5$). The pulse energies are 0.13 and 4.3 μJ for the propagating and driving pulses, respectively, whereas the waist is about 1.5 mm for the two beams. The pulse areas are then $\theta_p\simeq 0.3\pi$, $\theta_d=2.1\pi$ and the Rayleigh length is about 9 m, much larger than the length of the heat pipe (12 cm). The temperature inside the heat pipe was set to $T\simeq 145$ °C for which $e_{\mathrm{disp}}\simeq 3.4$. Experimental results are shown in Figs. 2 and 3. The spectrum intensity of the two-pulse sequence is displayed on a semilogarithmic scale. Both the theoretical and experimental curves in Fig. 2 exhibit a lateral band frequency symmetric to the propagating pulse spectrum and that has much smaller amplitude. It corresponds to the transitions between levels 3 and 2 in Fig. 1(b) for which is a small inversion of population occurs that has been neglected in the previous analysis (but not in the numerical simulations). In other respects, the dominant feature comes from the asymmetric distortion of the transmitted propagating pulse spectrum, as expected from the above theoretical analysis. Figure 3 shows the differential spectrum $S_p(1,W)-S_p(0,W)$ between the entrance and the exit $[S_p(Z,W)\propto|\hat{f}_p(Z,W)|^2]$ as a function of the wavelength and for which the asymmetry exhibits a dispersionlike shape. Numerical simulations results also exhibit very tiny oscillations near the atomic resonance which are not resolved experimentally by our spectrometer. These oscillations are also observed when measuring the transmitted spectrum intensity of a single strong pulse that propagates resonantly across the atomic medium [8,9] and involve the participation of optical ringing [5]. We focus next only on the interpretation of the asymmetry and we show that an analytical formula can be derived highlighting the underlying physics. Moreover, we consider the special case where the dispersion effects are important for the propagating pulse ($e_{\mathrm{disp}}\gg 1$) and the driving pulse is narrower than the propagating one ($\tau_{dp}<1$). We

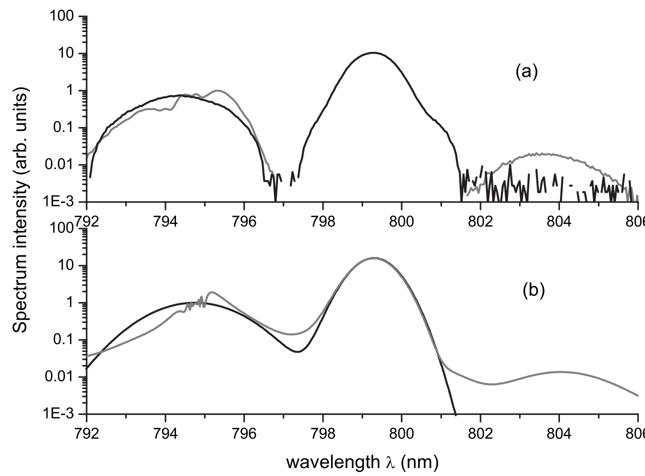

FIG. 2. Spectrum intensity of the two-pulse sequence. (a) Experiment. (b) Numerical simulations. In black is the spectrum at the entrance of the medium and in gray is the transmitted spectrum. See text for parameters values.

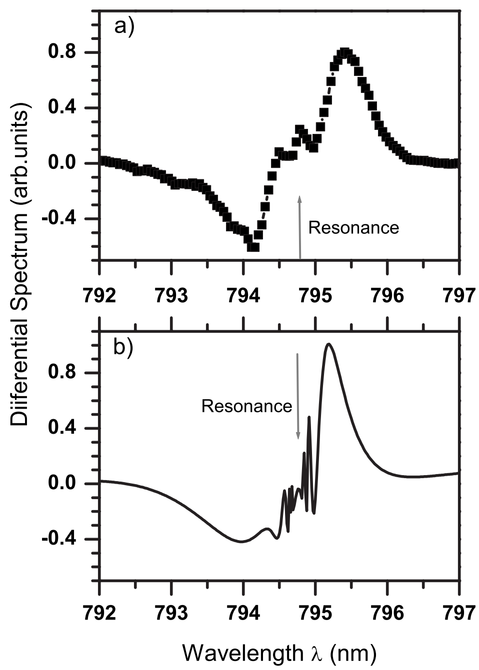

FIG. 3. Differential spectrum $S_p(1,W)-S_p(0,W)$ between the exit and the entrance of the sample in the region of the propagating pulse. (a) Experiment. (b) Numerical simulations. A dispersionlike behavior is observed.

assume that light-shift effects induce only a small perturbation on the propagation of the weak pulse e.g. $\hat{I}_s(0) \ll e_{disp}^{-1}(\ll 1)$. In this situation, equation (4) can be solved analytically for $|W| \gg e_{\text{disp}}/(4\hat{I}_s(0))$ leading to an effective diagonal response function (see Appendix B):

$$\text{Re}(W,W') \simeq ik\chi(W)\left(1 - i\frac{\hat{I}_s(0)}{2}\right)\delta(W-W') \qquad (5)$$

Integration of the propagation equation (4) then gives the following expression for the transmitted spectrum $S_p(1,W) \propto |\hat{f}_p(1,W)|^2$:

$$S_p(1,W) \simeq S_p(0,W)\{1 + k\hat{I}_s(0)R[\chi(W)]\}. \qquad (6)$$

Here $R[\chi]$ design the real part of χ. From Eqs. (5) and (6), the spectral components are decoupled and the system can be regarded as a stationary medium with a modified spectral response that induces a linear gain proportional to the adiabatic phase. The transmitted spectrum exhibits an asymmetry with amplification of red-detuned frequencies and absorption of blue-detuned components. This asymmetry reveals the asymptotic part of the linear susceptibility (we have $R[\chi(W)] = -e_{\text{disp}}/2kW$) that is imprinted directly on the spectrum. An interesting feature in this phenomenon is that although the lightshifts push apart the two levels and increase their separation to the blue, they influence both the blue and red-detuned frequency components of the propagating pulse by modifying their intensity weights. This is possible only because the transient character of the lightshifts. Indeed, if the driving pulse is monochromatic, the interaction reduces to a constant shift of the energy levels (1) and (4). In this case, a displacement of the absorption line from $W_d+\Delta$ to $W_d+(\Omega-\Delta)$ for the propagating pulse is the only effect that appears when recording the transmitted spectrum of the latter. Another interesting feature is that lightshifts induce a redistribution of the pulse energy without a neat gain for the propagating pulse energy [in the limit of the condition of application of relation (6)]. This result is in line with our initial assumptions, since level 3—for which an inversion of population with respect to level 2 occurs—is not excited by the propagating pulse and no population transfer due to the strong pulse occurs between the levels in the adiabatic approximation. Therefore, no energy can thus be exchanged between the pulses. If a long pulse (quasimonochromatic) propagates through the medium and its frequency is red-detuned, the inequality $|\int_{-\infty}^{T} f_p^{(0)}(Z,T')dT'| \ll 1$ does not hold and relation (5) does not remain valid, and as a consequence no amplification can be expected. In our experiment, $\hat{I}_s(0) \simeq 0.4$, $e_{\text{disp}}^{-1} \simeq 0.33$ and $\tau_{dp} \simeq 1.5$ so that the conditions $\hat{I}_s(0) \ll e_{\text{disp}}^{-1}$ and $\tau_{dp} < 1$ required for the strict application of the analytical formula (6) are not fulfilled in the experimental situation. Nevertheless, the dispersionlike behavior of the asymptotic part of the spectrum is still dominant, even if the $1/W$ dependence is perturbed by high-order terms that were neglected. The tiny oscillations observed in Fig. 3 appear in a spectral region where the validity of our analytical formula derived in Eq. (6) is not satisfied and as a consequence they are not predicted by our analytic treatment. These oscillations are thus the consequence of more complex reshaping effects when the above approximations no longer hold.

Finally, the situation described here (small lightshifts) can be seen as a particular case of the dynamic Kerr effect [14] in an original situation where the medium is optically thick and exhibits a noninstantaneous response due to the transient character of the lightshifts.

IV. CONCLUSION

We have shown that a two-level system driven by a strong nonresonant ultrashort pulse can be regarded for a weak resonant propagating pulse (with $\tau_{dp} < 1$) as a stationary atomic medium with a modified spectral response. The evolution has to be adiabatic ($\Delta \gg 1$), the dispersion significant and dominant with respect to the lightshift effects $[\hat{I}_s(0) \ll e_{\text{disp}}^{-1} \ll 1]$. The asymptotic part of the transmitted spectrum maps out the linear susceptibility function. An analytical formula has been derived and the dispersionlike feature observed experimentally. By highlighting the role of the lightshifts, these results give a new insight into the understanding of spectral reshaping of ultrashort pulses in the situation of a transient excitation. They pave the way to possible applications in laser spectroscopy for the probe of the long-range dispersion function of a complex system. Finally, the understanding of energy redistribution between the spectral components also represents an important step towards the challenge of the design of active pulse shapers that use propagation effects in order to amplify or create the spectral components [4,15].

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APPENDIX A

Using the Schrödinger equation, relations (1) and (2), the amplitudes $\alpha_+^{(1)}$ to the first order in respect with the propagating pulse can be written as

$$\alpha_+^{(1)}(Z,T) = -ie^{-i\int_{-\infty}^{T}[(\Delta-\Omega)/2]d(T'/\tau_{dp})} \times \int_{-\infty}^{T} V_{+-}(Z,T')e^{-i\int_{T'}^{T}\Omega d(T''/\tau_{dp})}dT', \quad \text{(A1)}$$

with $V_{ij}=\langle i|V^{(+)}+(V^{(+)})^{\dagger}|j\rangle$, $V^{(+)}=(\theta_p/2)(f_p e^{-i\Delta T/\tau_{dp}}|a\rangle\langle b|)$. Using the relation $\rho_p \simeq \alpha_-^{(0)^*}\alpha_+^{(1)}$ and Eqs. (3) and (A1), the envelope of the radiated field (per length unit) reduces to

$$\frac{\partial f_p}{\partial Z}(Z,T) = \int_{-\infty}^{+\infty} f_p(Z,T')R(T,T')dT', \quad \text{(A2)}$$

where $R(T,T')$ is the non-stationary time response function given by

$$R(T,T') = -\frac{e_{\text{disp}}}{2}h(T-T')L(T')L(-T), \quad \text{(A3)}$$

with $L(T)=e^{i\int_{-T}^{T}[I_s(T')/2]d(T'/\tau_{dp})}$, $I_s(T)=\Omega(T)-\Delta$, and $h(T-T')$ is the Heaviside function ensuring the causality of the interaction [with $h(T-T')=1$ if $T>T'$, $h(T-T')=0$ else] and which is responsible for the appearance of dispersion effects. The spectral behavior of the radiated field can be understood by performing a Fourier transform of relation (A2). With $W=(\omega-\omega_0)\tau_p$, we then obtain the relation (4) with the spectral response of the system

$$\text{Re}(W,W') = ik(2\pi)^{-2}[\chi(W)\hat{L}(W-W')] \otimes \hat{L}(-W). \quad \text{(A4)}$$

Here, $\hat{f}(W)=\int_{-\infty}^{+\infty}f(T)e^{iWT}dT$ denotes the Fourier transform of f and $\otimes$ is the convolution operation defined as $(f\otimes g)(x) =\int_{-\infty}^{+\infty}f(y)g(x-y)dy$. $\chi(W)=i(e_{\text{disp}}/2k)\hat{h}(W)$ is the linear susceptibility of the system with $k=k_pL/2$, $\hat{h}(W) =\lim_{\Gamma\to 0}[1/(\Gamma-iW)]=\pi\delta(W)+i\,\text{P}(1/W)$, where δ and P denote the Dirac function and the Cauchy principle value, respectively.

APPENDIX B

We use the relation $\int_{-T}^{T}I_s(T)dT=2\int_{-\infty}^{T}I_s(T)dT -\int_{-\infty}^{\infty}I_s(T)dT$. If $\hat{I}_s(0)=\int_{-\infty}^{\infty}I_s(T)d(T/\tau_{dp})\ll 1$, expand $L(T) =e^{i\int_{-T}^{T}[I_s(T')/2]d(T'/\tau_{dp})}$ to the first order and perform a Fourier transform operation. We get

$$\hat{L}(W) \simeq 2\pi\delta(W) - \hat{I}_s(W)\text{P}(1/W). \quad \text{(B1)}$$

Using the relation $\tau_{dp}<1$ and Eq. (B1), relation (4) can be expressed as

$$\frac{\partial \hat{f}_p}{\partial Z}(Z,W) \simeq \frac{ik}{2\pi}\Bigg(2\pi\chi(W)\hat{f}_p(Z,W) - \chi(W)\hat{I}_s(0)\text{P}\int_{-\infty}^{+\infty}\frac{\hat{f}_p(Z,W')}{W-W'}dW' + \hat{I}_s(0)\text{P}\int_{-\infty}^{+\infty}\frac{\chi(W')\hat{f}_p(Z,W')}{W-W'}dW'\Bigg). \quad \text{(B2)}$$

Using the relation $(i/2\pi)\text{P}\int_{-\infty}^{+\infty}[\tilde{\psi}(W')/(W'-W)]dW' =\int_0^{+\infty}\psi(T)e^{iWT}dT-[\tilde{\psi}(W)/2]$ and the fact that $|\int_{-\infty}^{T}f_p^{(0)}(Z,T')dT'|\ll 1$ for $e_{\text{disp}}\gg 1$, it follows that at the first order with respect to the lightshift phase $\hat{I}_s(0)$ and for frequencies W such as $|k\chi(W)\hat{I}_s(0)/2|\ll 1$ we have

$$\frac{\partial \hat{f}_p^{(1)}}{\partial Z}(Z,W) \simeq ik\left(\chi(W)f_p^{(1)}(Z,W) - \frac{i}{2}\chi(W)\hat{I}_s(0)\hat{f}_p^{(0)}(Z,W)\right). \quad \text{(B3)}$$

The following relation:

$$\frac{\partial \hat{f}_p}{\partial Z}(Z,W) \simeq ik\left(\chi(W)f_p(Z,W) - \frac{i}{2}\chi(W)\hat{I}_s(0)\hat{f}_p(Z,W)\right) \quad \text{(B4)}$$

holds with $\hat{f}_p=\hat{f}_p^{(0)}+\hat{f}_p^{(1)}$, leading to the effective diagonal response function given by relation (5).

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