

Modeling the subpicosecond pulse compression in the vacuum ultraviolet and the extreme ultraviolet

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A method for pulse compression to the subpicosecond range in the VUV and the XUV is investigated theoretically. The induced phase modulation on a signal by a pump pulse near two-photon resonance in inert gases is analyzed, including group-velocity dispersion, pulse walk-off, initial delay, shift of the signal peak, and linear and nonlinear chirp rates for Gaussian and hyperbolic secant pulses. A reasonable agreement is found with the theoretical and experimental results of other authors.

INTRODUCTION

The techniques for ultrashort-pulse generation and compression have been extensively developed in recent years. Shorter pulses permit more accurate timing measurements and response investigations of certain atomic and molecular systems to higher frequencies. Several groups reported on the generation of high-power pulses of subpicosecond duration using an excimer laser as a last stage.^{1,2} Pulses of 43-fs duration are obtained by intracavity frequency doubling in a colliding-pulse mode-locked laser at $\lambda = 310$ nm.³ Ultrashort pulses in the XUV and x-ray range would permit investigations with high temporal resolution in solid-state and plasma physics.^{4,5} Time compression of x-ray pulses from electron-storage rings using equitemporal optics and x-ray pulse lengths down to 100 fs seems feasible.⁶

Because of the relatively low peak powers of the existing coherent sources in the VUV and the XUV, self-phase modulation (SPM) does not offer a possibility to obtain enough chirping for pulse compression. Alfano and co-workers reported that intense picosecond pulses could be used to enhance the spectral broadening of weaker pulses, copropagating in bulk glasses.^{7,8} Since then, several groups have been studying cross-phase modulation and induced phase modulation (IPM) effects.⁹⁻¹⁵

Recently, we have modeled the compression of nanosecond UV pulses from excimer lasers and Raman-shifted pulses using IPM.¹⁶

In this paper we report an extension of this method to the subpicosecond range in the VUV and the XUV ranges.

INDUCED PHASE MODULATION IN THE VUV AND XUV

Our calculations of the nonlinear susceptibility for SPM, $\chi_{\text{IPM}}^{(3)}$, in inert gases in the VUV and the XUV¹⁷ have shown that the existing coherent sources^{18,19} with relatively low peak power can induce a chirp with a bandwidth that is insufficient for compression of pulses in the nanosecond and picosecond ranges. The length of the waveguides used is limited for technical reasons to several centimeters.²⁰

High values of the nonlinear susceptibility for IPM, $\chi_{\text{IPM}}^{(3)} = \chi^{(3)}(\omega_S; \omega_S, \omega_P, -\omega_P)$, can be obtained if one

chooses the wavelengths of the signal, λ_S , and the pump, λ_P , in the vicinity of a two-photon resonance. If the wavelength pair is chosen in such a way that $\omega_S + \omega_P > \omega_{\text{res}}$ (ω_{res} is the resonance frequency), the nonlinear susceptibility $\chi_{\text{IPM}}^{(3)}(\omega_S)$ can be kept negative, thus avoiding the induced focusing of the signal. Since the two-photon resonances in inert gases are high lying, λ_S and λ_P can be selected far from single-photon resonances, which reduces the absorption. Table 1 shows the lowest two-photon resonances in inert gases, the respective resonant-level energy ΔE , and a possible wavelength pair (λ_S, λ_P) having a high nonlinear susceptibility. The signal wavelength is selected to coincide with VUV and XUV sources reported in the literature, and the respective pump wave can be obtained by the widely used dye laser and other lasers. It should be noted that, by a fixed detuning from resonance, $\chi_{\text{IPM}}^{(3)}$ decreases from Xe I to He I.¹⁷

The resonant character of $\chi_{\text{IPM}}^{(3)}$, however, possesses bandwidth limits. This intrinsic limitation becomes important at two-photon detuning from resonance of the order of the chirp bandwidth. The spectral broadening $\Delta\lambda_S$ of a transform-limited compressed pulse with a fixed duration α_S decreases to shorter wavelengths λ_S . This dependence is shown in Fig. 1 (curve 1). The other dependence shown (curve 2), the nonlinear susceptibility $|\chi_{\text{IPM}}^{(3)}|$, strongly increases with small detunings. A compressed pulse with a certain duration can be obtained through nonlinearity with a maximum value, determined by a detuning from resonance not smaller than the expected spectrum broadening of the signal (Fig. 1). This plot can be used for an estimate of the required length of the nonlinear medium. From the other side a strong limit to this length could be posed by the group-velocity mismatch. Further, the response time of the nonlinearity is especially important, since it affects chirping of subpicosecond pulses.

INDUCED PHASE MODULATION IN A DISPERSE NONLINEAR MEDIUM

In our model, signal pulses I_S and pump pulses I_P are traveling in a hollow waveguide containing a medium with cubic nonlinearity and density N . The present considerations are made by the following restrictions: a negli-

Table 1. Nonlinear Medium, Two-Photon Resonance, Transition Energy, and a Possible Signal- and Pump-Wavelength Pair, Ensuring High Values of the Nonlinear Susceptibility for Pump-Induced Phase Modulation

Gas	Two-Photon Resonance	ΔE (eV)	λ_S (nm)	λ_P (nm)
He I	1s-2s	20.615	64	1054
Ne I	$2p^6-3p[1/2]$	18.382	83	351
	$2p^6-3p[3/2]$	18.694		
Ar I	$3p^6-4p[1/2]$	12.907	117	535
	$3p^6-4p[3/2]$	13.283		
Kr I	$4p^6-5p[1/2]$	11.304	157	364
	$4p^6-5p[3/2]$	12.100		
Xe I	$5p^6-6p[1/2]$	9.580	193	393
	$5p^6-6p[3/2]$	10.958		

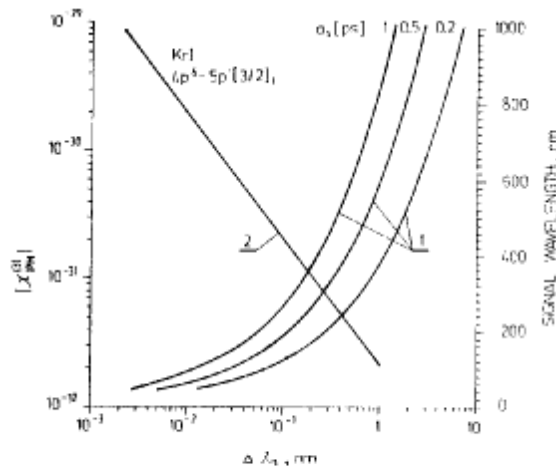


Fig. 1. Selection chart for IPM and compression of optical pulses. Curves 1: spectral broadening $\Delta\lambda_S$ versus wavelength for fixed compressed-signal pulse duration. Curve 2: nonlinear susceptibility versus detuning from two-photon resonance.

gible SPM, a negligible IPM of the pump from the signal ($I_S \ll I_P$), a coordinate system connected with the signal, a negligible efficiency of the four-wave mixing and parametric processes, and, finally, a fast nonlinear response with respect to the pulse duration.

The Schrödinger equation (SE), describing the evolution of the slowly varying signal pulse, is

$$i \frac{\partial \psi_S}{\partial x} + \alpha_S \frac{\partial^2 \psi_S}{\partial \tau^2} + k^{IPM}(\omega_S) |\psi_S|^2 \psi_S = 0. \quad (1)$$

The input pulses have a Gaussian form:

$$\psi_S(x, \tau) = A_S(x) \exp[-\tau^2/2a_S^2(x) + i b_S(x) \tau^2], \quad (2a)$$

$$\psi_P(x, \tau) = A_P(x) \exp[-(\tau - \tau_D + x v_{SP})^2/2a_P^2(x)], \quad (2b)$$

where $a_S(x)$ and $a_P(x)$ are the pulse half-widths at the $1/e$ level, $b_S(x)$ is the frequency chirp parameter, $A_S(x)$ and $A_P(x)$ are the complex amplitudes, τ_D is the initial delay between the pulses, and $v_{SP} = |v_{GS}^{-1} - v_{GP}^{-1}|$ is determined by the group-velocity mismatch. In Eq. (1), $\alpha_S = (-1/2)(\lambda_S^3/2\pi c^2)(\partial^2 n/\partial \lambda^2)_{\lambda=\lambda_S}$ is the group-velocity

dispersion (GVD) coefficient and $k^{IPM}(\omega_S)$ is the nonlinear coefficient, i.e.,

$$k^{IPM}(\omega_S) = [3\pi^2 N \chi_{IPM}^{(3)}(\omega_S)] [\lambda_S n_0^2(\omega_S)]^{-1}.$$

Using the variational functional L corresponding to the SE,²¹

$$L = \frac{i}{2} \left(\psi_S \frac{\partial \psi_S^*}{\partial x} - \psi_S^* \frac{\partial \psi_S}{\partial x} \right) + \alpha_S \left| \frac{\partial \psi_S}{\partial \tau} \right|^2 - k^{IPM}(\omega_S) |\psi_S|^2 |\omega_P|^2, \quad (3)$$

we obtain the following differential equations for the pulse duration $a_S(x)$ and the chirp rate $b_S(x)$:

$$\frac{da_S}{dx} = 4\alpha_S a_S b_S, \quad (4)$$

$$a_S \frac{db_S}{dx} = -4\alpha_S a_S b_S^2 + \frac{\alpha_S}{a_S^3} - k^{IPM}(\omega_S) |A_P|^2 \times \frac{a_P/a_S}{(a_S^2 + a_P^2)^{3/2}} \exp\left[-\frac{(\tau_D - x v_{SP})^2}{a_S^2 + a_P^2}\right] \times \left[1 - \frac{a_P^2}{a_S^2 + a_P^2} - \frac{2a_S^2(\tau_D - x v_{SP})^2}{(a_S^2 + a_P^2)^2}\right]. \quad (5)$$

It is important to evaluate how well the variational approach can take into account the combined effects of IPM, pulse walk-off, and initial delay.^{9,10} If the pump pulse has an initial delay with respect to that of the signal, the latter sees, in a medium with a normal dispersion, the leading front of the pump pulse, and its spectral distribution is shifted to the red side of the spectrum. Conversely, with no initial delay, the signal pulse feels the trailing front of the pump, the spectral shift is toward the blue, and, when the pump pulse passes completely through that of the signal, the IPM does not change the spectral distribution.⁹

We have compared the results of our approach with the theoretical and experimental results of Baldeck et al.⁹ We show in Fig. 2 the induced wavelength shift versus the

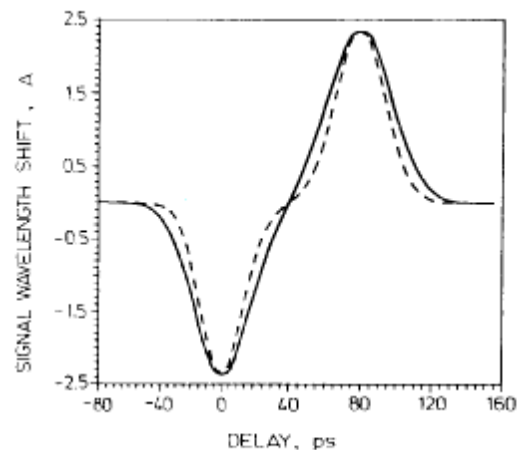


Fig. 2. Comparison between the variational method result (solid curve) and the result of Ref. 9 (dashed curve) in the case of signal-wavelength shift versus initial delay between the input pulses ($\lambda_S = 532$ nm, $\lambda_P = 1064$ nm, $a_P = \sqrt{2} a_S = 19.8$ ps, length of the optical fiber $L = 1.04$ m, i.e., four times the walk-off length).

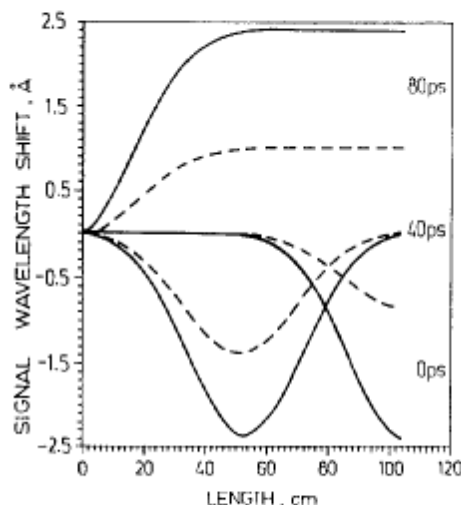


Fig. 3. Influence of the medium losses on the signal-wavelength shift. The conditions are the same as those in Fig. 2.

Initial delay, calculated for the experimental conditions of Ref. 9. A reasonable agreement is seen for both dependencies. When the GVD is neglected, Eq. (4) can be integrated directly in order to obtain an expression for the chirp bandwidth, similar to that in Ref. 9 but normalized to $(a_s^2 + a_p^2)^{1/2}$ instead of a_p (i.e., T_0). This result describes adequately the case of the induced linear chirp at $a_p \gg a_s$, certainly with a reduced compression ratio.^{12,22}

The influence of the medium losses due to single- and two-photon absorption can be accounted for by a damping nonlinear coupling in the SE:

$$k^{IPM}(\omega_s) \rightarrow k^{IPM}(\omega_s, x) = k^{IPM}(\omega_s) \exp(-2\gamma x), \quad (6)$$

where γ is the decay constant. Figure 3 plots the variation of the induced wavelength shift over the length of the medium for fixed initial delay without (solid curves) and with (dashed curves) pump losses in the medium. As one can see from the figure, the effect of the absorption on the induced wavelength shift is a minimum for pulses fully overlapping at the entrance of the medium.

The variational approach used here permits the selection of a different probe function to describe the signal and pump pulses. In Appendix A we show the results obtained for a hyperbolic secant type of pulse.

PUMP-INDUCED STEEPENING AND NONLINEAR CHIRP

An important condition for a successful pulse compression by a pair of gratings is the absence of nonlinear frequency chirp. Let us now consider the propagation of signal and pump pulses, accounting for the rates of the linear (b_s) and nonlinear (c_s) chirp and the temporal shift of the signal peak, originating in the intensity dependence of the group velocity of the signal pulse.²³ For simplicity of presentation, we have neglected the influence of the initial delay between the pulses and the group-velocity mismatch v_{SP} , accounted for in the previous model. We have retained the GVD α_s , although v_{SP} is more significant than α_s . The analysis is done in a single moving coordinate system. The equation describing the IPM on a probe pulse from an intense pump pulse traveling together is²¹

$$i \frac{\partial \psi_s}{\partial x} + \alpha_s \frac{\partial^2 \psi_s}{\partial \tau^2} + k^{IPM}(\omega_s) |\psi_p|^2 \psi_s + i\beta \frac{\partial}{\partial \tau} (|\psi_p|^2 \psi_s) = 0, \quad (7)$$

where $\beta = 2k^{IPM}(\omega_s)/\omega_s$. The additional nonlinear term in Eq. (7) as compared with Eq. (1) arises when the slowly varying envelope approximation must be amended to account for short-pulse propagation. We assume Gaussian input pulses:

$$\psi_s(x, \tau) = A_s(x) \exp[-(\tau - \xi)^2/2a_s^2(x) + ib_s(x)\tau^2 + ic_s(x)\tau^3], \quad (8a)$$

$$\psi_p(x, \tau) = A_p(x) \exp[-\tau^2/2a_p^2(x)]. \quad (8b)$$

It can be shown that the Lagrangian

$$L = \frac{i}{2} \left(\psi_s \frac{\partial \psi_s^*}{\partial x} - \psi_s^* \frac{\partial \psi_s}{\partial x} \right) + \alpha_s \left| \frac{\partial \psi_s}{\partial \tau} \right|^2 - k^{IPM}(\omega_s) |\psi_s|^2 |\psi_p|^2 + i\beta |\psi_p|^2 \psi_s \frac{\partial \psi_s^*}{\partial \tau}, \quad (9)$$

when it is inserted into the Lagrange equation, gives Eq. (7). Using the above variational procedure, we obtain a system of ordinary differential equations for the variational parameters (see Appendix B). These results need some comment.

Since no initial assumptions are made for b_s and c_s , we can treat them as complex parameters. The imaginary parts b_s^i and c_s^i , which can be either positive or negative, can be interpreted as small corrections to the signal pulse duration:

$$A_s = A_{s0} \exp[-(i\tau - \xi)^2/2a_s^2 + b_s^i \tau^2 + c_s^i \tau^3 + i(b_s^r \tau^2 + ic_s^r \tau^3)], \quad (10)$$

Neglecting for a moment the GVD and getting $\xi \ll a_s$ and $a_s = a_p$, we obtain, from Eq. (B2) below,

$$\xi = 2^{-1/2} \beta |A_p|^2 x. \quad (11a)$$

Following the approach of Anderson and Lisak,²³ applied to the SPM we obtained²⁴

$$\xi = \beta |A_p|^2 x. \quad (11b)$$

In the limiting case in which β , ξ , and c_s are neglected, the results, given in Appendix B, reproduce our previous analyses.¹⁶

Let us illustrate the method on the example of $\lambda_s = 121.6$ nm, the Ly- α transition of the hydrogen atom, owing to the special interest in the generation of coherent emission in this wavelength.¹⁴ An appropriate gas is Kr 1, where $\omega_s + \omega_p$ is near the two-photon resonance $4p^2 - 5p[3/2]_1$ at $\lambda_p = 650$ nm. If a duration of the compressed pulse of the order of 500 fs is desirable, we obtain from Fig. 1 a minimum detuning from resonance, $\Delta\lambda = 4 \times 10^{-2}$ nm, and a respective nonlinear coefficient, $|\chi_{IPM}^{(3)}| \leq 3 \times 10^{-31}$ esu. Our estimates are for $\Delta\lambda = 0.1$ nm, from which $\chi_{IPM}^{(3)}(\omega_s) = -1.4 \times 10^{-31}$ esu. The waveguide considered is an aluminum capillary with an internal diameter of $2\rho = 200$ μ m, filled with 1 atm Kr 1. The calculated group-velocity mismatch is $v_{SP} = 6.4$ ps/cm and, at input pulses of 30 ps (FWHM), the length of the nonlinear medium is limited to $L = 3$ cm. The GVD co-

efficient is calculated to be $\alpha_S = -9.1 \times 10^{-27} \text{ s}^2/\text{cm}$. The intensity of the pump pulse is taken as $I_P = 2 \times 10^{11} \text{ W/cm}^2$ ($P_P = 6.3 \times 10^5 \text{ W}$), considerably smaller than the critical value for self-focusing ($P_{\text{critical}}^{\text{SF}} = 1.3 \times 10^7 \text{ W}$). The second model predicts a compression ratio of $F = 140$ ($F = a_S \Delta\omega^{\text{IPM}}/2$, where $\Delta\omega^{\text{IPM}}$ is the chirp bandwidth). The first model, which includes the group-velocity mismatch between λ_S and λ_P , gives a compression ratio of $F = 72$ for the signal ($a_S^{\text{MIN}} = 420 \text{ fs}$). This reduction of the proposed signal compression efficiency results from the group-velocity mismatch and the pulse walk-off¹⁰ and is to be expected in a potential experiment. The shift of the signal peak is $\xi = 32 \text{ fs}$, which is negligible compared with the initial duration $a_S(x=0) = 30 \text{ ps}$. The ratio $c_S^R a_S/b_S^R \leq 10^{-3}$ shows that the nonlinear chirp would not influence considerably the pulse compression. The corrections to the pulse duration from b_S^I and c_S^I are of the order of 1 fs and can be neglected with respect to a_S and a_P .

The nonlinear susceptibilities are calculated numerically according to the single-sided Feynman diagrams.²⁵ The full expression for $\chi_{\text{IPM}}^{(3)}$ consists of 24 terms. In the vicinity of $\omega_S + \omega_P$, the two-photon resonance $\chi_{\text{IPM}}^{(3)}$ is determined mostly by terms of the type

$$\frac{r_{GA}^2 r_{AF}^2}{\omega_{GF} - \omega_S - \omega_P - i\Gamma} \{ (\omega_{GA} - \omega_S)^{-2} + (\omega_{GA} - \omega_P)^{-2} + [(\omega_{GA} - \omega_P)(\omega_{GA} - \omega_S)]^{-1} \}, \quad (12)$$

where r_{ij} denotes the corresponding dipole matrix element and Γ is the damping constant of the transition. The matrix elements for bound-bound²⁶ and bound-free²⁷ transitions are corrected for sign.^{28,29} Within the accuracy of the matrix elements used, the accuracy of the nonlinear susceptibility calculation is estimated to be 40%.

The high negative value of $\chi_{\text{IPM}}^{(3)}(\omega_S)$ is indicative of induced defocusing. In the case considered here the refractive index of the aluminum capillary is³⁰

$$n_{\text{Al}}(\lambda_S = 121.6 \text{ nm}) = 0.607,$$

and the maximum decrease of the refractive index of the nonlinear medium was calculated to be

$$n(\lambda_S) = n_0(\lambda_S) + (1/2)n_{\text{IPM}}^{\text{NL}}(\lambda_S)|A_P|^2 = 0.995.$$

Therefore the waveguide mode of propagation will not be significantly disturbed by the induced defocusing of the signal. The characteristic length for multimode structure formation in the waveguide,³¹

$$z_0 = [\rho/(2\theta_c)] \exp(V/2), \quad (13)$$

where $\theta_c = \arccos[n_{\text{Al}}/n(\lambda_S)]$ is the critical angle and $V = (2\pi/\lambda_S)\rho n(\lambda_S)\sin(\theta_c)$ is the waveguide parameter, is more than 2 orders of magnitude larger than the waveguide length considered, and hence the forms of Eqs. (1) and (7) are correct.

CONCLUSION

We have analyzed the inert gases as a possible nonlinear medium for compression of ultrashort pulses in the VUV and the XUV. The lowest-lying two-photon resonances in

He I, Ne I, Ar I, Kr I, and Xe I ensure high nonlinear coefficients for pump-induced modulation when pump and signal pulses copropagate along the medium. The variational method is shown to describe adequately the theoretical and experimental results of other authors. The compression of pulses to the subpicosecond range can be optimized with respect to wavelength detuning from resonance and medium length. The initial delay between the pulses and the group-velocity mismatch are shown to influence negatively the pulse compression. Our analyses show that the variational method for solving the Schrödinger equation is an appropriate description of Gaussian, hyperbolic secant, and other types of functions as well as the temporal shift of the signal peak and the velocities of the signal and pump pulses. In a model experiment, pulses as short as 420 fs on the Ly- α transition at 121.6 nm of the hydrogen atom seem to be obtainable by input pulses of 30 ps. A numerical solution of the corresponding equations can show, however, how well the phase-modulated signal compresses as a function of the experimental parameters proposed.

APPENDIX A

The Schrödinger equation and the Lagrangian do not depend on the specific choice of the probe function. Let the signal and pump pulses be approximated by a hyperbolic secant function, namely,

$$\psi_S(x, \tau) = A_S(x) \text{sech}[\tau/a_S(x)] \exp[i b_S(x) \tau^2], \quad (A1)$$

$$\psi_P(x, \tau) = A_P(x) \text{sech}[(\tau - \tau_D + x v_{SP})/a_P(x)]. \quad (A2)$$

Solving the reduced variational problem, we obtain a system of two ordinary differential equations:

$$\frac{da_S}{dx} = 4\alpha_S a_S b_S, \quad (A3)$$

$$a_S \frac{db_S}{dx} = -4\alpha_S a_S b_S^2 + \frac{4\alpha_S}{\pi^2 a_S^3} + \frac{6k^{\text{IPM}}(\omega_S) |A_P|^2}{\pi^2} \times \left\{ -\frac{B}{a_S A^2} + \frac{B a_P \cosh^2[(\tau_D - x v_{SP})/a_P]}{a_S^2 A^2} - \frac{a_P \sinh^2[(\tau_D - x v_{SP})/a_P]}{a_S^2 A} \right\}, \quad (A4)$$

where

$$A = \cosh^2[(\tau_D - x v_{SP})/a_S] \cosh^2[(\tau_D - x v_{SP})/a_P] - 1, \quad (A5)$$

$$B = \sinh^2[(\tau_D - x v_{SP})/a_P] \sinh[2(\tau_D - x v_{SP})/a_S] \times [(\tau_D - x v_{SP})/a_S]. \quad (A6)$$

When the initial delay and the group-velocity mismatch are negligible, Eq. (A4) is transformed into

$$a_S \frac{db_S}{dx} = -4\alpha_S a_S b_S^2 + \frac{4\alpha_S}{\pi^2 a_S^3} + \frac{6k^{\text{IPM}}(\omega_S) |A_P|^2 a_P}{(a_S^2 + a_P^2)^2} \times (a_P^2 - a_S^2 - 2a_S a_P). \quad (A7)$$

APPENDIX B

The system of the coupled ordinary differential equations for the variational parameters a_S , ξ , b_S^R , b_S^I , c_S^R , and c_S^I

has the form

$$\frac{da_s}{dx} = [\alpha_s/(2\xi^2 - a_s^2)](6b_s^R a_s \xi^2 - 4b_s^R a_s^3 - 12c_s^R a_s \xi^3 + 36c_s^R a_s \xi^3) + [1 - D/(2\xi^2 - a_s^2)] \times [2\beta a_r^2 \xi / (a_s a_s^2 + a_r^2)^{3/2}] C, \quad (B1)$$

$$\frac{d\xi}{dx} = [\alpha_s/(a_s^2 - 6\xi^2)](9c_s^R a_s^4 + 4b_s^R a_s^2 \xi - 12c_s^R \xi^4 - 8b_s^R \xi^3) + DC3\beta a_r^2 / [(6\xi^2 - 3a_s^2) \times (a_s^2 + a_r^2)^{3/2}], \quad (B2)$$

$$\frac{db_s^R}{dx} = -[\alpha_s/a_s^4(2\xi^2 - a_s^2)]\{2\xi^2 + a_s^2 + [(c_s^R)^2 - (c_s^I)^2] \times (36\xi^4 a_s^4 - 27a_s^6) + 4a_s^4(2\xi^2 - a_s^2) \times [(b_s^R)^2 - (b_s^I)^2]\} + [C/(2\xi^2 - a_s^2)] \times \{h^{(M)}(\omega_s)[-2\xi^2(2\xi^2 + 2a_s^2) - a_r^2(2\xi^2 - a_s^2) + a_s^4](a_s^2 + a_r^2)^{-3/2} + 2\beta_s^R a_r^2 E - c_s^R F\}, \quad (B3)$$

$$\frac{db_s^I}{dx} = -[\alpha_s/a_s^4(2\xi^2 - a_s^2)]\{-2c_s^R c_s^I(36\xi^4 a_s^4 + 27a_s^6) + 8a_s^4 b_s^R b_s^I(2\xi^2 - a_s^2)\} + [C/(2\xi^2 - a_s^2)] \times (\beta E + 2\beta b_s^I a_r^2 E - c_s^I F), \quad (B4)$$

$$\frac{dc_s^R}{dx} = [4\alpha_s/3(2\xi^2 - a_s^2)]\{\xi a_s^{-4} + 9(c_s^R b_s^R - c_s^I b_s^I) a_s^2 - 36(c_s^R)^2 \xi^3 + 36(c_s^I)^2 \xi^3 - 18(c_s^R b_s^R - c_s^I b_s^I) \xi^2\} + [4C/3a_s(2\xi^2 - a_s^2)]\{2h^{(M)}(\omega_s)\xi^3 a_s \times (a_s^3 + a_r^2)^{-3/2} + \beta b_s^R a_r^2 G - \beta c_s^R H\}, \quad (B5)$$

$$\frac{dc_s^I}{dx} = [4\alpha_s/3(2\xi^2 - a_s^2)]\{9(c_s^R b_s^I - c_s^I b_s^R) a_s^2 - 72\xi^3 c_s^R c_s^I - 18(c_s^R b_s^I - c_s^I b_s^R) \xi^2\} + [4C/3a_s(2\xi^2 - a_s^2)] \times (\beta G/2 + \beta b_s^I a_r^2 G - \beta c_s^I H), \quad (B6)$$

where

$$C = |A_r|^2 a_r \exp[-\xi^2/(a_s^2 + a_r^2)], \quad (B7)$$

$$D = 4\xi^2 - a_s^2[1 + 2\xi^2 a_r^2 / (a_s^2 a_s^2 + a_r^2)], \quad (B8)$$

$$E = (-5\xi a_s^4 - 2\xi a_r^4 - 7\xi a_s^3 a_r^2 - 2\xi^3 a_r^2 + 4\xi^5) \times (a_s^2 + a_r^2)^{-3/2}, \quad (B9)$$

$$F = [3\beta a_r^2 / 2a_s^2(a_s^2 + a_r^2)^{3/2}]\{44\xi^2 a_s^5 a_r^4 + 19\xi^2 a_s^2 a_r^6 + 14\xi^4 a_s^4 a_r^2 + 32\xi^4 a_s^2 a_r^4 - 6\xi^4 a_r^6 - 12\xi^4 a_s^6 - 24\xi^4 a_s^2 a_r^2 + 6a_s^6 a_r^4 + 7a_s^4 a_r^6 + 17\xi^2 a_s^6 a_r^2 - 8\xi^2 a_s^5 + a_s^{10}\}, \quad (B10)$$

$$G = a_s[(a_s^2 + a_r^2)^3 - 4\xi^4 + 4\xi^2(a_s^2 + a_r^2)] \times (a_s^2 + a_r^2)^{-3/2}, \quad (B11)$$

$$H = [3a_r^2 \xi / 2a_s(a_s^2 + a_r^2)^{3/2}]\{-3a_s^2 a_r^4(a_s^2 + a_r^2) - 5\xi^2 a_s^4 a_r^2 - 6\xi^2 a_s^2 a_r^4 + \xi^2 a_r^6 + 2\xi^2 a_s^6 + 4\xi^4 a_s^2 a_r^2\}. \quad (B12)$$

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