Enhancement of third-harmonic generation by Stark-chirped rapid adiabatic passage

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Received 11 July 2003; received in revised form 4 September 2003; accepted 5 September 2003

Abstract

We report enhancement of third-harmonic generation in a nonlinear optical medium, prepared in maximum coherence by Stark-chirped rapid adiabatic passage (SCRAP). In this technique a strong off-resonant infrared radiation pulse induces an adiabatic passage process by sweeping the transition frequency of a two-photon transition in Krypton atoms through resonance with a near-resonant pump radiation pulse at 213 nm, appropriately delayed with respect to the infrared radiation. During the adiabatic passage process, a transient two-photon maximum coherence is induced, which beats with the pump laser frequency. Detrimental effects of Doppler broadening and collisional dephasing in the medium are reduced during the SCRAP process. The efficiency of third-harmonic generation, yielding vacuum-ultraviolet radiation at 71 nm is enhanced by more than one order of magnitude with respect to the case of conventional frequency conversion in an unprepared medium.

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PACS: 42.50.–P; 42.65.–k; 32.80.–t; 42.50.–Gy; 42.50.–Hz

1. Introduction

Efficient generation of vacuum-ultraviolet (VUV) radiation is a topic of considerable scientific efforts, as it is of significant interest for many applications, e.g. laser lithography, high resolution microscopy or spectroscopy. Frequency conversion processes like third-harmonic generation or four-wave mixing, usually applied in rare gases or metal vapours, have therefore been investigated thoroughly in the last decades [1]. Due to the rather small third-order nonlinear susceptibility, such conventional techniques suffer from relatively poor conversion efficiencies, typically in the range of $10^{-6}$–$10^{-4}$. If resonances are used to enhance the nonlinear optical response, re-absorption of the generated radiation strongly limits the attainable efficiency.

Coherent preparation of the nonlinear optical medium may serve to overcome these difficulties. Absorption can be suppressed by the well-known
phenomenon of electro-magnetically induced transparency (EIT) [2,3]. EIT may be applied, e.g. to enhance the efficiency of four-wave mixing processes involving atomic resonances. A pump laser drives a two-photon transition between an atomic ground state \( |1\rangle \) and an excited state \( |2\rangle \) at frequency \( \omega_{12} \). In turn radiation from a dressing laser, tuned to the transition between the excited state \( |2\rangle \) and a state \( |3\rangle \) at frequency \( \omega_{23} \), is mixed with the pump radiation field to yield radiation at \( 2\omega_{12} \pm \omega_{23} \). If the dressing laser-induced transition is strongly and coherently driven, the system may be prepared in a dark state, which allows the suppression of resonance absorption by destructive quantum interference, while frequency conversion is still permitted due to the non-vanishing third-order susceptibility.

As a consequence EIT reduces both the linear as well as the nonlinear optical response. In contrast the concept of maximum coherence can be applied to significantly enhance the nonlinear response.

Consider a two-level system of a ground and an excited state: The polarisation of the medium, which determines the efficiency of any frequency conversion process, depends upon the amplitude and phase of the coherence between the ground and the excited state [5]. For an equal amplitude, coherent superposition of the ground and the excited state the coherence reaches a maximum. Such a maximum coherence can be prepared in a robust way by adiabatic passage techniques. The atomic coherence acts now like an oscillator with a maximum amplitude. If a probe radiation field is introduced, the beat frequencies of this oscillator with the probe radiation field are generated with high conversion efficiencies.

Frequency conversion processes in systems prepared under conditions of maximum coherence have been investigated both theoretically [4,8,15] and experimentally [9–13]. Frequency upconversion close to the vacuum-ultraviolet spectral region involving maximum coherence has been implemented in lead vapour [12] using a technique closely related to stimulated Raman adiabatic passage (STIRAP) [19]. In this setup the ground state \( |1\rangle \) of a lambda-type level scheme is coupled to state \( |2\rangle \) by a pump laser pulse of frequency \( \omega_{12} \), the intermediate state to state \( |3\rangle \) by the Stokes laser pulse at frequency \( \omega_{23} \). The Stokes pulse has to precede the pump pulse (which can be automatically arranged in an optically dense medium [12]) in order to provide adiabatic evolution of the system, therefore driving all the population from the ground state \( |1\rangle \) to the target state \( |3\rangle \) [19]. During the process a maximum coherence, driven at frequency \( \omega_{12} \pm \omega_{23} \) is produced transiently. An additional radiation field can be used to beat with this maximum coherence. Such, radiation at 185 nm was obtained by Harris and coworkers [12] in an efficient frequency conversion process.

The structure of a lambda-type system prepared by STIRAP does not permit generation of vacuum-ultraviolet radiation with much shorter wavelength, because the maximum coherence is driven at rather small frequency \( \omega_{12} \pm \omega_{23} \). Multi-photon transitions could extend the accessible wavelength regime, but the Stark shifts, which are intrinsic to the strong fields required for multi-photon excitations prevent adiabatic population transfer by the STIRAP technique [6].

To overcome these difficulties and to apply the concept of maximum coherence to systems involving multi-photon excitations, thus enabling generation of vacuum-ultraviolet radiation far below 200 nm, we previously suggested to use the technique of Stark-chirped rapid adiabatic passage (SCRAP) [15,16]. The SCRAP technique permits efficient adiabatic population transfer in coupling schemes even involving multi-photon excitations and may also be applied to Doppler broadened media [14,19].

In the implementation of SCRAP discussed here a ground state \( |1\rangle \) and an excited state \( |2\rangle \) are coupled near (but not exactly on) resonance by a two-photon transition, induced by a pump laser pulse at frequency \( \omega_{12} \). An additional off-resonant laser pulse induces dynamic Stark shifts, which drive the atomic system through resonance with the pump laser. Provided the laser pulses are appropriately delayed with respect to each other, all of the population is driven from the ground to the excited state in a rapid adiabatic passage process. As for STIRAP, a maximum coherence is established during the SCRAP process at the point when half of the population is driven to the
excited state whilst the other half remains in the ground state. The system is then prepared in a transient state of maximum coherence and acts as a local oscillator, driven at the two-photon transition frequency $2\omega_{12}$. The temporal behaviour of the coherence can be controlled by the pulse delay and static detuning of the pump laser pulse. An additional radiation field at frequency $\omega_3$ can be used to beat with the maximum coherence and generate radiation fields at frequencies $2\omega_{12} \pm \omega_3$. If the pump laser field itself is used instead of an additional field, the four-wave mixing process degenerates to third-harmonic generation, yielding short-wavelength radiation at frequency $3\omega_{12}$ with high conversion efficiency.

We like to stress, that a transient maximum coherence can also be provided without the Stark shifting laser, if the detuning of the pump laser frequency from resonance is larger than the natural linewidth and the evolution of the system is adiabatic. In this case population is driven from the ground state to the excited state in the first half of the pump pulse and returns coherently to the ground state in the second half of the pulse. This process is known as coherent population return (CPR). Although no net population can be detected in the excited state after the interaction, a transient coherence builds up during the process. This situation is similar to that pertaining to maximal vibrational coherence generation in recently reported Raman schemes where a small detuning (of the order of the Doppler width) from Raman resonance ensures all the molecules are excited into the correctly phased state [13]. The bandwidth of CPR is determined by the driving Rabi frequency [7]. Thus, if the Rabi frequency is smaller than the Doppler width of the medium, CPR will not work efficiently.

While techniques for the preparation of maximum coherence other than SCRAP suffer from Doppler and collisional broadening as discussed above, such perturbing effects are easily reduced in the SCRAP process. Provided the laser-induced Stark shift is larger than the bandwidth of Doppler broadening, all atoms in the medium are driven in maximum coherence. Therefore the nonlinear optical medium can be used in the most efficient way.

2. Atomic coupling scheme

We have chosen Krypton atoms as the non-linear optical medium to implement the preparation of a maximum coherence by SCRAP. The coupling scheme, as depicted in Fig. 1, is as follows: The ground state $4p^6\,^1S_0$ and the excited state $4p^5\,^5P_{1/2}$ (lifetime $\tau = 20$ ns [17]) in Krypton are coupled by a pump laser pulse at $\lambda_p = 213$ nm slightly detuned from two-photon resonance. The Stark shifts, driving a rapid adiabatic passage process, are induced by a second radiation pulse at $\lambda_{St} = 1064$ nm, far off any atomic resonance.

![Fig. 1. Coupling scheme. A pump laser pulse at 213 nm couples the atomic ground state $4p^6\,^1S_0$ and the excited state $4p^5\,^5P_{1/2}$ in Krypton. The pump laser frequency is slightly tuned off two-photon resonance. A second laser pulse at 1064 nm provides dynamic Stark shifts, driving the transition frequency through resonance with the pump laser and inducing a rapid adiabatic passage process. During the process a transient maximum coherence is prepared in the system, which beats with the fundamental pump laser frequency and significantly enhances third-harmonic generation of the pump radiation field to yield vacuum-ultraviolet radiation at 71 nm.](image-url)
The coherence established at twice the pump laser frequency beats with the fundamental frequency of the pump laser and enhances the generation of the third-harmonic at $\lambda = 71$ nm. We compare the efficiency of this frequency conversion process, supported by maximum coherence, with respect to conventional third-harmonic generation, when the pump laser is tuned to exact two-photon resonance and the Stark shifting laser is switched off.

3. Experimental setup

The experimental setup is as follows: Krypton atoms in natural abundance are expanded in a supersonic jet with stagnation pressure of typically 300 mbar through a pulsed nozzle (general valve, opening diameter 0.8 mm). The laser pulses are focused into the atomic jet 1 mm behind the nozzle orifice. The atomic density in the interaction region is estimated to be some $10^{18}$ cm$^{-3}$.

The pump laser pulse is provided by the frequency-tripled output of a pulsed dye amplifier (PDA, Quanta Ray). The amplifier is seeded by typically 500 mW cw emission from a tunable, single-mode cw dye laser (Coherent 699) at 638 nm and pumped by the second harmonic frequency of a pulsed, injection-seeded Nd:YAG laser (GCR 4, Quanta Ray). Because both the cw seed as well as the pulsed Nd:YAG laser provide single-mode radiation, the output of the pulsed dye amplifier is a radiation pulse with transform-limited bandwidth and centre frequency determined by the cw seed laser. The visible output of the amplifier, providing pulse energies up to 15 mJ, is collimated by a telescope system and frequency doubled in a BBO crystal. The polarisation of the ultraviolet radiation at 319 nm is rotated by a half-wave plate and mixed with the remaining visible radiation at 638 nm in a second BBO crystal to obtain a laser pulse in the far-ultraviolet spectral region at $\lambda_P = 213$ nm with a pulse duration of $\tau_P = 3.6$ ns (FWHM of intensity) and a pulse energy of up to 1.5 mJ. A fused silica Pellin–Broca prism is used to spectrally disperse the visible and ultraviolet radiation pulses. The laser pulse at 213 nm passes an optical delay line and is focused with a quartz lens ($f = 175$ mm) into the atomic beam. The beam waist in the interaction region is 30 μm (FWHM) providing laser intensities up to the GW/cm$^2$ regime.

Residual infrared radiation from the ND:YAG pump laser at $\lambda_{St} = 1064$ nm with a pulse energy of 180 mJ and a pulse duration of $\tau_{St} = 9.9$ ns (FWHM) serves as the Stark shift inducing radiation pulse. The infrared radiation is focused by a lens ($f = 215$ mm) and combined with the ultraviolet pump laser radiation by a dichroic mirror. The beam waist of the infrared laser in the interaction region is about 350 μm (FWHM), yielding peak intensities of approximately 12 GW/cm$^2$. The confocal parameters of the laser beams were measured to be approximately 1 cm, thus substantially longer than the length of the interaction region, which is about 1 mm. Therefore inside the medium a variation of the laser intensities in the propagation direction may be considered as negligible.

A VUV-spectrometer (Model VM-502, Acton research, arm-length 20 cm), equipped with an iridium coated spherical grating ($f = 10$ cm, 1200 grooves/mm), serves to separate the fundamental laser frequencies at 213 and 1064 nm from the vacuum-ultraviolet radiation, generated by frequency conversion in the atomic jet. Besides the third-harmonic frequency of the pump radiation at 71 nm we observed four-wave mixing of the ultraviolet pump pulse and the infrared Stark shifting laser pulse, yielding radiation at frequencies $2 \omega_P \pm \omega_{St}$, corresponding to wavelengths of 97 nm and 118 nm. All of these nonlinear optical processes occur simultaneously, but they do not compete with each other. The sum-frequency mixing signal could be used to pre-align the spatial overlap of the laser foci in the experiment. For optimised sum-frequency mixing the laser foci should not be shifted with respect to each other. Therefore the atoms, which are excited by the pump laser will experience the peak intensity in the infrared laser focus. In contrast, the efficiency of third-harmonic generation assisted by SCRAP depends critically upon the laser-induced Stark shift, which was subsequently varied by shifting the infrared laser focus over the smaller pump laser focus.

After passing the spectrometer, the vacuum-ultraviolet radiation is detected in an electron multiplier tube (Hamamatsu, type R595). The
output current of the electron multiplier is amplified in a fast broadband amplifier, integrated in a boxcar averager (SRS, model SR250) and processed in a PC.

4. Results and discussion

Figs. 2(a)–(c) show the relative intensity variation of the vacuum-ultraviolet radiation at 71 nm as a function of the pump laser frequency varied in the range of the two-photon resonance. For each of the Figs. (a)–(c) the relative intensity was normalised to the peak value of the CPR case, when the Stark shifting laser was switched off. Figs. (a)–(c) show the results for different values of the pump laser peak intensity and delay between the laser pulses (defined with respect to the peak intensities of pump and Stark laser pulse). The intensity of the pump radiation was (a) \( I_p = 380 \text{ MW/cm}^2 \), (b) \( I_p = 5.7 \text{ GW/cm}^2 \) and (c) \( I_p = 180 \text{ MW/cm}^2 \) while the delay between the two laser pulses was (a, b) \( \Delta \tau = 7.6 \text{ ns} \) and (c) \( \Delta \tau = 12 \text{ ns} \), with the Stark laser pulse preceeding the pump pulse. The lower and upper trace in each of the graphs (a)–(c) indicate the case of the infrared laser switched off or on, respectively. The insets show enlarged views for better visibility.

When the infrared laser is switched off, conventional resonantly enhanced third-harmonic generation is observed (see Figs. 2(a)–(c) lower traces). Due to the quite low pump laser intensity of 380 MW/cm\(^2\) and 180 MW/cm\(^2\), the VUV yield and hence the signal-to-noise ratio in the lower traces in Figs. 2(a) and (c) is rather poor. The third-harmonic intensity (see Fig. 2(a), lower trace and inset) reaches a maximum around two-photon resonance. The width of the two-photon resonance is about 4–5 GHz (FWHM) as it

Fig. 2. Experimental results. Relative intensity of vacuum-ultraviolet radiation at 71 nm, obtained by third-harmonic generation of the pump radiation field, when the frequency of the pump laser frequency is varied and the Stark shifting laser is switched off (lower traces and insets to the left) or on (upper traces and insets to the right). The pump laser intensity is (a) 380 MW/cm\(^2\), (b) 5.7 GW/cm\(^2\) and (c) 180 MW/cm\(^2\). The delay between the two laser pulses is (a, b) \( \Delta \tau = 7.6 \text{ ns} \) and (c) \( \Delta \tau = 12 \text{ ns} \). In each graph the data are normalised with respect to the peak intensity of the generated vacuum-ultraviolet radiation for the case of the Stark shifting laser switched off. Without the Stark shifting laser, conventional third-harmonic generation is observed (lower traces). For better visibility the structure is enlarged in the insets. When the Stark shifting laser is switched on, an enhancement of 22 is observed in the conversion efficiency (a). The maximum of the resonance is blue-shifted with respect to the case of conventional frequency conversion. For higher intensities (b) or larger pulse delay (c) the optimum enhancement is reduced.
is mainly determined by the Doppler broadening in the atomic beam, which is estimated to be about 3 GHz (with respect to the one-photon detuning). Additional broadening may be due to collisions in the medium. For the given pump laser intensity the Rabi frequency is estimated to be $\Omega_p = 2.4 \text{ ns}^{-1} \approx 190 \text{ MHz}$ (with respect to one-photon detuning), which is substantially smaller than the Doppler width. Thus, as discussed above, the conversion process strongly suffers from inhomogeneous broadening, even if the system is driven in a coherent population return process.

A more detailed look on the lineshape reveals a dip in the centre of the resonance, which is due to nonadiabatic evolution involving ionisation losses. The dip can be used to calibrate the frequency axis to the exact position of the two-photon resonance.

When the Stark shifting laser is switched on and introduced into the system 7.6 ns prior to the pump pulse, a significant enhancement by a factor of 22 is observed in the efficiency of third-harmonic generation (see Fig. 2(a), upper trace). In contrast to conventional frequency conversion (see Fig. 2(a), lower trace) the maximum intensity is obtained for the pump laser frequency detuned by 3 GHz from two-photon resonance.

The resonance is broadened to about 8 GHz (FWHM) which clearly exceeds the Doppler width of the medium and shows an asymmetric lineshape, as it is typical for the SCRAP process [14]. Only for positive (static) detunings $\Delta\nu_p$ of the pump laser frequency the atomic system is driven through resonance by the (infrared) laser-induced dynamic Stark shifts. Therefore the blue-shifted asymmetric lineshape indicates the sign of the dynamic Stark shift to be positive, which coincides with our estimations below.

The absolute value of the peak Stark shift $S$ can be estimated from the condition $0 \leq 2\Delta\nu_p \leq S$, which determines the range of pump laser detunings, permitting the resonance condition to be met in the SCRAP process during the interaction with the pump laser. Thus, from the data in Fig. 2(a) we conclude the peak Stark shift to be $S \approx 200 \text{ ns}^{-1} \approx 16 \text{ GHz}$ (with respect to one-photon detuning), which is more than an order of magnitude smaller than the possible peak Stark shift, as it can be calculated from the atomic parameters (see below) to be $S_{\text{max}} = 5500 \text{ ns}^{-1} \approx 430 \text{ GHz}$ for the given infrared laser intensity (see above). The experimentally determined Stark shift is large enough to compensate for Doppler broadening in the medium, therefore additionally supporting the frequency conversion process in the coherently prepared medium.

The discrepancy between the observed and calculated Stark shift mirrors the fact, that the spatial overlap between the pump and the Stark shifting laser was experimentally aligned in a way to obtain the most appropriate Stark shift for the preparation of the maximum coherence in the interaction region (see above). Because the focus of the infrared laser was much bigger than the focus of the pump laser, it was possible to maintain good spatial overlap, while shifting the gaussian profile of the infrared laser focus across the interaction region. Thus the intensity in the interaction region was not necessarily the same as the possible peak intensity in the infrared laser focus, but could be substantially smaller.

In order to investigate the dependence of the conversion efficiency upon the pump laser intensity we have varied the pump pulse energy while all other experimental parameters were kept fixed (see Figs. 2(a) and (b)). We found, that the enhancement for third-harmonic generation decreases continuously with increasing pump laser intensity. Fig. 2(b) shows the intensity of the generated vacuum-ultraviolet radiation for a rather high pump laser intensity of $I_p = 5.7 \text{ GW/cm}^2$ ($\Omega_p = 35.5 \text{ ns}^{-1} \approx 2.8 \text{ GHz}$). The enhancement compared to conventional third-harmonic generation is merely 2, although, on the first glance, SCRAP is expected to be even more stable at higher intensities. In contrast, the strongest enhancement for frequency conversion was found for quite moderate laser intensities, i.e. $I_p = 380 \text{ MW/cm}^2$ (see Fig. 2(a)).

This can be understood regarding the effect of inhomogeneously broadening on the frequency conversion process. Without the Stark shifting laser, the system can be prepared in maximum coherence by CPR, thus by appropriate detuning from exact two-photon resonance [13]. The spectral (homogeneous) width of this maximum coherence is determined by the Rabi frequency $r_p$ by power broadening. If this width is smaller than the
Doppler width, as it is the case for small pump laser intensities, a SCRAP-induced maximum coherence, combined with SCRAP-induced compensation of inhomogeneous broadening, strongly enhances the frequency conversion process. In contrast, for high pump laser intensities, even without the Stark shifting laser, the width of the power broadened maximum coherence exceeds the Doppler width, thus compensates inhomogeneous broadening. In this case SCRAP cannot enhance the frequency conversion process any further. The enhancement drops for higher laser intensities. For a Rabi frequency clearly exceeding the Doppler width the observed enhancement would be expected to approach unity asymptotically. Residual enhancement may originate from averaging effects based on a gaussian pump laser intensity distribution. Only in the intense centre of the beam the Doppler width may be compensated by power broadening, while the intensity in the wings is too small. Thus, SCRAP offers some residual enhancement even for higher intensities.

In our experiment the width of the resonance for the case of the Stark shifting laser switched off (see lower trace in Fig. 2(b)) is about 8.8 GHz, thus clearly exceeding the Doppler width. This is confirmed by the estimated Rabi frequency $\Omega_p = 35.5\text{ns}^{-1} \approx 2.8$ GHz, which is approaching the Doppler width. Therefore the linewidth remains almost unchanged, when the Stark shifting laser is switched on (Fig. 2(b), upper trace and inset to the right) and the observed enhancement by SCRAP is small.

The two-photon Rabi frequency for the $4p^6 \quad ^1S_0 - 4p^5 5p \mid 01/2 \rangle_0$-transition in Krypton is calculated taking only the intermediate state $4p^5 5s \mid 11/2 \rangle_1$, which is closest to one-photon resonance (see Fig. 1) and likely to give the largest contribution, into account. With the transition dipole moments for the one-photon $4p^6 \quad ^1S_0 - 4p^5 5s \mid 11/2 \rangle_1$- and $4p^5 5s \mid 11/2 \rangle_1 - 4p^5 5p \mid 01/2 \rangle_0$-transitions [17] and using the general expression [5]

$$\Omega_p^{(2)} = \sum_i \frac{\mu_{1i}\mu_{2i}}{2\hbar^2 A_{1i}} E^2,$$

we obtain for the two-photon Rabi frequency of the $4p^6 \quad ^1S_0 - 4p^5 5p \mid 01/2 \rangle_0$-transition

$$\Omega_p^{(2)} \text{ (ns}^{-1}) \approx 6.2 \cdot I_p \text{ (GW/cm}^2\text{)}.$$

Using the expression for the dynamic Stark shift of the excited state $\mid 2 \rangle$ with respect to the ground state $\mid 1 \rangle$ by off-resonant coupling to a manifold of states $\mid i \rangle$ [5]

$$S = -\frac{1}{\hbar^2} \sum_i \left( \frac{\left| \mu_{2i} \right|^2}{4A_{2i}} - \frac{\left| \mu_{1i} \right|^2}{4A_{1i}} \right) E^2,$$

including all intermediate atomic states in Krypton up to $5p^5 \quad 6d \mid 11/2 \rangle$ and taking the transition dipole moments from [17] the estimated net dynamic Stark shift by the infrared laser yields

$$S_{St} \text{ (ns}^{-1}) \approx 455 \cdot I_{St} \text{ (GW/cm}^2\text{)}.$$

The Stark shifts of the much weaker pump laser pulse are negligible with respect to the Stark shifts of the infrared laser and ignored in the following.

Finally, photoionisation losses of the upper state $4p^5 \quad 5p \mid 01/2 \rangle_0$ by the pump laser are calculated with the ionisation cross section [18] $\sigma = 3$ Mb, to yield an ionisation rate of

$$\Gamma_2 \text{ (ns}^{-1}) \approx 4.3 \cdot I_p \text{ (GW/cm}^2\text{)}.$$

This indicates strong photoionisation of the sample at laser intensities in the GW/cm$^2$ regime.

The numerical simulations in Fig. 3 illustrate the efficiency of third-harmonic generation in a Doppler-broadened medium, coherently prepared either by CPR or SCRAP. The numerical algorithm is based on density matrix calculations for the population and coherence dynamics in the medium and integration of the propagation equations for the electric fields [15,16]. The figure shows the calculated efficiency for third-harmonic generation for the case of Rabi frequencies $(a, c) \quad \Omega_p = 3$ ns$^{-1}$ and $(b, d) \quad \Omega_p = 20$ ns$^{-1}$, with the pump laser frequency varied in the range of the two-photon resonance. The amplitude of the Stark-shift was chosen to be $S_0 = 200$ ns$^{-1}$. With the given experimental parameters the condition for adiabatic evolution in the SCRAP-scheme yields $\Omega_p > 4$ ns$^{-1}$ [14]. The Stark-shift by the pump laser, ionisation losses and spontaneous decay of the upper level population were neglected in the numerical calculations. The phase mismatch due to resonances outside the two-level system has been estimated [21] to be
\[ \Delta k = 2 \times 10^3 \text{ m}^{-1}, \] which corresponds to a phase mismatch per atom of \[ C = \Delta k / N = 2 \times 10^{-16} \text{ cm}^2. \] Due to these approximations and assumptions, the numerical calculation presented here can only be expected to give an order-of-magnitude estimation with respect to absolute frequency conversion efficiencies. Nevertheless, they serve to discuss the experimental results at least qualitatively in a very appropriate way.

The solid lines in Fig. 3 indicate the conversion efficiency for the Stark shifting laser switched off (CPR), the dashed lines for the Stark shifting laser switched on (SCRAP). The lower two graphs (c) and (d) show a convolution of the calculated efficiencies with a gaussian profile \[ \text{width 5.3 GHz (FWHM with respect to one-photon detuning)}, \] e.g. resulting from Doppler broadening. While for the lower Rabi frequency (a) the calculated linewidth in the case of CPR is 120 MHz – thus significantly smaller than the Doppler width – the linewidth for the case of SCRAP already is 3.5 GHz. SCRAP-assisted third-harmonic generation yields about the same peak intensity as in the case of CPR. After convoluting both datasets (a) and (b) with the Doppler profile, the efficiency of SCRAP-assisted third-harmonic generation shows an enhancement by 12 with respect to CPR. In contrast, when the Rabi frequency is increased to \[ \Omega_p = 20 \text{ ns}^{-1} \] (b), the third-harmonic linewidth for the case of CPR is increased to 2.3 GHz, thus approaching the Doppler width. For SCRAP the third-harmonic spectrum shows a broad contribution and a narrow feature, which exceeds the efficiency of the system prepared by CPR. The nature of the small feature is a permanent maximum coherence when the static detuning from resonance is very large. In this case adiabaticity for full population transfer by SCRAP is no longer provided. The transfer efficiency is reduced to 50% and a permanent maximum coherence is established instead of the transient coherence prepared by CPR. The convolution with the Doppler profile (d) yields almost no enhancement by SCRAP, as expected.
The numerical simulations differ by a factor of less than two to the SCRAP-induced observed enhancement of third-harmonic generation and clearly support the experimental observations. Remaining deviations to the experimental data are mainly due to the fact, that no integration over the spatial pump laser profile was performed in the calculations (see above). Additional effects by ionisation, dephasing by electron collisions [20] may also modify the process.

The effect of inhomogeneous broadening, as discussed above, is also mirrored in the dependence of the conversion efficiency upon the delay between the pump and Stark shifting laser. Fig. 2(c) shows the third-harmonic intensity for a delay of \( \Delta \tau = 12 \) ns, increased by 4.5 ns with respect to the data shown in Fig. 2(c). The pump laser intensity was \( I_p = 180 \) MW/cm\(^2\), which resulted in the highest enhancement of conversion efficiency. While the enhancement of the SCRAP experiment with respect to the CPR case was about 22 for a delay of \( \Delta \tau = 7.6 \) ns (Fig. 2(a)), it reduces to four for this larger delay of \( \Delta \tau = 12 \) ns. At the same time the third-harmonic linewidth for the SCRAP case reduces to 2.5 GHz, which is about the Doppler width. The additional delay diminishes the Stark shift at the time of the peak of the pump pulse by a factor of 11, leading to an expected linewidth of 800 MHz, which is essentially smaller than the Doppler width. Thus, the smaller Stark shift results in a reduced enhancement, as the homogeneous linewidth can no longer compensate for Doppler broadening.

We also investigated the effect of the pulse order on the conversion efficiency. We found that the peak conversion efficiency remains almost unchanged if the Stark laser pulse either preceeds or follows the pump laser pulse. The efficiency of the population transfer in a SCRAP-driven system is expected to be the same in the one or other pulse sequence [14]. Thus the induced coherence as well as the efficiency of frequency conversion is also expected to remain unchanged. The pulse sequence solely causes a change in the observed lineshape, but barely affects the peak conversion efficiency.

We like to note, that the preparation of a maximum coherence, either by SCRAP or by CPR, is only possible for excitation by radiation pulses with transform-limited bandwidth. For conventional laser systems, providing laser pulses with much broader bandwidth, the frequency conversion process will be less efficient and adiabatic preparation of the medium is not possible due to phase fluctuations on the pump laser pulse.

5. Conclusions

We demonstrated strong enhancement of third-harmonic generation in a nonlinear optical medium, prepared in maximum coherence by Stark chirped rapid adiabatic passage. In this coupling scheme, a pump laser at \( \lambda_p = 213 \) nm drives a two-photon transition between the atomic ground state \( 4p^6 \, ^1S_0 \) and the excited state \( 4p^5 \, 5p \, ^1P_0/2 \) in Krypton. The frequency of the pump laser is slightly detuned from exact two-photon resonance. A second laser at \( \lambda_{St} = 1064 \) nm, delayed by about one pulse duration with respect to the pump laser pulse, induces dynamic Stark shifts and drives a rapid adiabatic passage process, preparing a transient coherent superposition of the ground and excited state in the medium. This maximum coherence as well as compensation of Doppler broadening by the laser-induced Stark shift serves to strongly enhance any subsequent frequency conversion process, provided the Stark shift is larger than the inhomogeneous broadening. For the case of moderate pump laser intensities in the regime of some 100 MW/cm\(^2\), the efficiency of third-harmonic generation to yield vacuum-ultraviolet radiation at 71 nm was enhanced by more than one order of magnitude with respect to conventional frequency conversion in the Doppler broadened medium. For higher pump laser intensities in the regime of several GW/cm\(^2\) and larger pulse delays the enhancement of SCRAP-assisted frequency conversion with respect to conventional techniques is still observable, but less significant.

In conclusion SCRAP offers major advantages for frequency conversion driven by moderate intensities, i.e. intensities which are just sufficient to saturate a relevant pump transition, in inhomogeneously broadened media. As in many implementations of frequency conversion to the regime of vacuum-ultraviolet radiation, the nonlinear
optical media are dense, i.e. Doppler broadened, and the pulse energy of the driving radiation is limited, SCRAP addresses an experimentally challenging situation.

Acknowledgements

We acknowledge support from the European Union, RTN contract number HPRN-CT-1999-000129, the Deutsche Forschungsgemeinschaft (DFG) as well as the German-Israeli Foundation (GIF), contract number I-644-118.5/1999. The authors like to thank K. Bergmann, University of Kaiserslautern for encouragement as well as valuable discussion and comments on the manuscript.

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