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LETTER TO THE EDITOR

## High-order tunable sum and difference frequency mixing in the XUV region

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**Abstract.** We present a study of tunable high-order sum and difference frequency mixing in the 7–70 eV range, in xenon, argon and neon. We mix the light from a terawatt, sub-picosecond titanium–sapphire laser and that from a tunable optical parametric generator. The radiation generated through mixing processes involves absorption or emission of one or two photons from the weak tunable source. The relative strengths of the sum and difference frequency processes depend strongly on the wavelength range of the generated radiation. The sum frequency processes are stronger at low photon energies in xenon, whereas the difference frequency processes dominate at high photon energies, in neon, being almost comparable to harmonic generation.

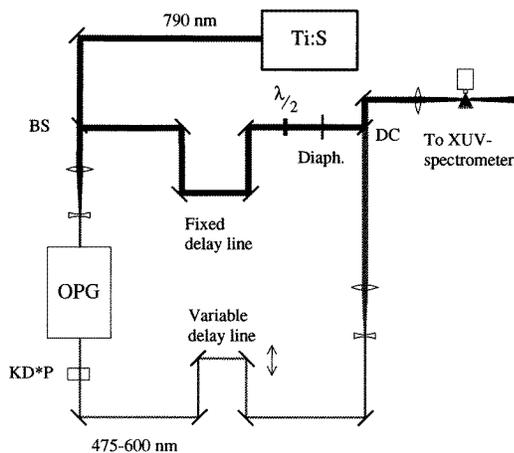
High-order sum and difference frequency mixing in rare gases has recently attracted much interest, both experimentally (Eichmann *et al* 1994, 1995, Perry and Crane 1993, Watanabe *et al* 1994) and theoretically (Eichmann *et al* 1995, Ivanov *et al* 1995, Long *et al* 1995). Most of the experiments have been performed by mixing the light from a high-power short-pulse laser and its second or third harmonic. Depending on the relative intensities and polarizations of the two fields, the odd harmonics can be considerably enhanced (Eichmann *et al* 1995, Watanabe *et al* 1994). In the experiments using the second harmonic, even harmonics of the fundamental light are generated with, in some cases, a strength almost comparable to that of the odd-order ones (Perry and Crane 1993). Moreover, the variation of the relative phase of the two fields allows one, in principle, to control harmonic generation in different ways (Ivanov *et al* 1995, Long *et al* 1995). Experiments have also been performed by mixing the light from an intense fixed-frequency laser with a tunable light source, leading to *tunable* radiation in the XUV region. This was first demonstrated by Eichmann and co-workers (1994), using light from a titanium sapphire (Ti:S) laser mixed with tunable radiation from an optical parametric generator (OPG). They generated tunable radiation up to  $\sim 31$  eV, with a tuning range of about 30% between consecutive harmonics.

There are several advantages of using an OPG for the frequency mixing as compared to using the second or third harmonic. One is, of course, the aspect of tunability, which is of crucial importance for applications. The other is the possibility to identify, and hence to study, sum versus difference frequency generation, since the two processes lead to

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different generated wavelengths when the two fields have non-commensurate frequencies. The relative importance of the different frequency conversion mechanisms depends both on the dynamics of an atom in the presence of two fields of different colours, and on phase matching, which makes a clear distinction between sum and difference frequency mixing (Shkolnikov *et al* 1993, 1995, Bjorklund 1975). On the other hand, since the frequencies of the two fields are non-commensurate, phase control is not possible in this type of experiment.

In this letter, we present a study of high-order sum and difference frequency mixing processes in rare gases, and, in particular, we investigate the difference between the two processes. The experiments are performed by mixing the light from a terawatt, sub-picosecond Ti:S laser and that from an OPG pumped by the Ti:S laser. We compare the efficiencies for these mechanisms in different rare gases, i.e. in different conditions of laser intensities and wavelength ranges.



**Figure 1.** Experimental set-up for sum and difference frequency mixing.

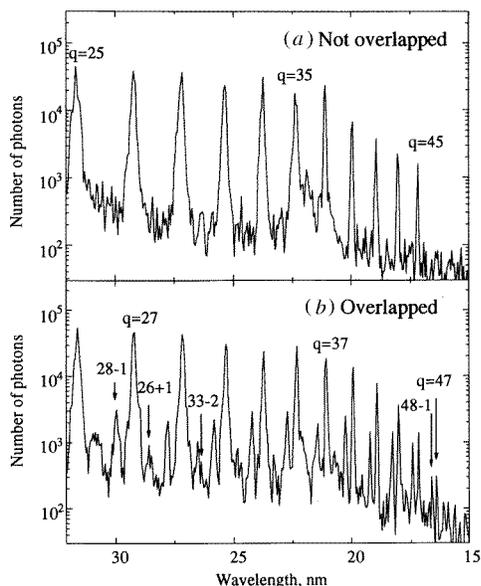
A schematic diagram of the experimental set-up is shown in figure 1. The essence of the set-up is the overlap in space and time of two beams of different colours. The Ti:sapphire laser system, of the DRECAM facility in Saclay, is based on chirped pulse amplification and generates 100 mJ, 150 fs pulses at 790 nm with a 20 Hz repetition rate. The beam is horizontally polarized and has a diameter of 4 cm. A pulse energy of 60–70 mJ is used in the present experiment. A beamsplitter, marked as BS in the diagram, directs 25% of this energy to pump the OPG (Danielius *et al* 1993), of the Lund high-power laser facility. A telescope just before the OPG reduces the beam diameter to 1 cm to increase the pump intensity and to match the size of the OPG optics. The non-linear medium in the OPG is a type II BBO ( $\beta$ -barium borate) crystal in which radiation from 1150 to 2700 nm can be produced, by angle tuning, in a three-pass process. Parametric superfluorescence is generated in a first step and is spectrally and spatially narrowed in a second pre-amplification step. The radiation generated is finally amplified in an energy boosting third step. The signal or idler output from the OPG is mixed with the fundamental radiation in a KD\*P crystal, and radiation in the 475–600 nm range can be generated by type II sum-frequency mixing. When the OPG is pumped by 10 mJ of infrared, the pulse energy of the generated light (green in most of the experiments) is approximately 0.4 mJ. The polarization is vertical.

A variable delay line is installed in the path of the green light to control the temporal overlap necessary for the high-order mixing experiment. A telescope increases the green beam to a diameter of  $\sim 5$  cm to decrease the size of the focal spot and hence increase the focused intensity. Moreover, it enables control of the divergence of the green beam,

which is different from that of the infrared one, and hence improves the spatial overlap in the focus. Well controlled focusing of the green light is difficult due to the limited spatial coherence of the OPG output. Further matching of the green and infrared spot sizes requires that a second telescope reduces the diameter of the infrared beam. This has been used in some of the measurements. The remainder of the pump beam is removed by a dichroic mirror with a 99.9% reflection at 790 nm, and the infrared signal and idler beams from the OPG are filtered away after a number of reflections of the green light in other dichroic mirrors.

The 75% of the infrared light not used to pump the OPG is strongly diaphragmed to a diameter of a few mm and a pulse energy of a few mJ, to maximize the efficiency for harmonic generation and frequency mixing. The polarization is rotated to vertical using a  $\lambda/2$  plate, since parallel polarizations are much more efficient for frequency mixing than perpendicular polarizations (Eichmann *et al* 1995, Perry and Crane 1993). The two beams, with parallel polarizations, are brought together by a dichroic mirror—indicated as DC in the figure. The beams are focused by the same  $f = 45$  cm lens into a jet of xenon, argon or neon gas. The intensities in the focus are estimated to be between  $10^{14}$  and  $10^{15}$  W cm $^{-2}$  for the infrared and  $10^{12}$ – $10^{13}$  W cm $^{-2}$  for the green light. The radiation generated in the gas jet is analysed with an XUV spectrometer with no entrance slit, utilizing a 700 lines/mm plane grating at grazing incidence and a toroidal mirror. The XUV light is detected with an electron multiplier.

Two spectra obtained in neon are shown in figure 2. The spectra are obtained by stepwise rotation of the grating, and each point is an average of 30 shots. In figure 2(a) the two beams are not overlapped in time—only in space—and hence only odd harmonics of the infrared light are observed with the plateau extending up to approximately  $q = 37$ , and with the highest harmonic  $q = 45$ . The intensity of the green light is not high enough to generate harmonics in this spectral range. The feature between the 35th and the 37th harmonic is due to incoherent plasma light. In figure 2(b) the two beams are overlapped in time as well, and several additional series of peaks are observed. The peak just to the right



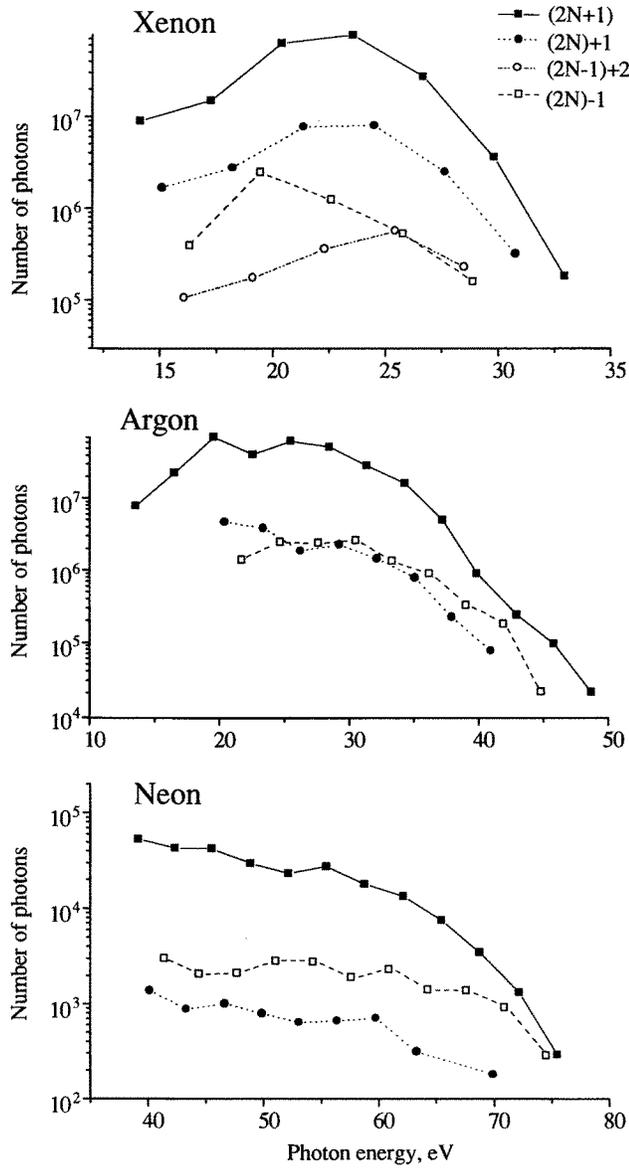
**Figure 2.** Two spectra obtained in neon. In (a) the two beams are only overlapped in space, whereas in (b) the beams are overlapped in both space and time. The indication  $q = 27$  refers to the 27th harmonic of the infrared, and  $28 - 1$  refers to the signal with frequency  $28\omega_{\text{IR}} - \omega_{\text{OPG}}$ .

of the harmonic signal of order  $(2N + 1)$  corresponds to a frequency of  $2N\omega_{\text{IR}} + \omega_{\text{OPG}}$  and will be referred to as a  $(2N) + 1$ . As in harmonic generation, parity conservation requires that the total number of photons absorbed or emitted by the two laser fields is odd. The peak just to the left corresponds to a frequency of  $(2N + 2)\omega_{\text{IR}} - \omega_{\text{OPG}}$  and will be referred to as  $(2N + 2) - 1$ . We will, in general, denote sum (respectively difference) frequency processes involving absorption (emission) of one photon from the OPG field by  $+1$  ( $-1$ ). Finally, peaks with frequencies of  $(2N + 3)\omega_{\text{IR}} - 2\omega_{\text{OPG}}$  (or, in general,  $-2$ ) are barely visible between the  $(2N - 2) + 1$  and the  $(2N + 2) - 1$  peaks. The mixed signals extend out to  $48 - 1$ , which corresponds to a wavelength of 17 nm or a photon energy of 73 eV. The last harmonic is now  $q = 47$ , and when comparing the two spectra, the cut-off seems to be less steep when the green light is present. This is the case in several spectra in neon and argon. Note that the strength of the  $-1$  signals is significantly larger than the  $+1$  signals, and is even approaching that of the harmonic signals at high energies.

In argon, up to the  $32 - 1$  signal has been observed, with a wavelength of 26 nm, or a photon energy of 48 eV, and in xenon up to the  $20 - 1$  signal. The efficiency for mixed frequency generation increases from neon to argon, and from argon to xenon, as is the case for harmonic generation. The mixed signals are typically a factor of 10–50 weaker than the corresponding harmonic peaks in the plateau, but are almost comparable in the cut-off region. In xenon, peaks with frequencies  $(2N - 1)\omega_{\text{IR}} + 2\omega_{\text{OPG}}$  have been observed, with a strength approximately one order of magnitude lower than the  $+1$  signals.

Tunability is probably the most interesting aspect of frequency mixing for future applications. The present experimental results demonstrate tunability up to 70 eV, which extends the previous results of Eichmann *et al* (1994) quite far into the XUV. With the present tuning range of the OPG, and including  $\pm 2$  processes, i.e. in xenon, 70% of the spectrum between two subsequent harmonics can be covered, apart from a small region close to the harmonics. At shorter wavelengths, in argon or neon, where ‘two-photon’ processes are barely detectable, about 40% of the range can be covered with the  $\pm 1$  signals, leaving an additional gap centred between the two peaks. However, ‘two-photon’ signals could probably be enhanced in argon and neon as well with a higher OPG energy, which could be obtained by pumping harder. The tuning range could be further extended to cover the entire spectrum between two harmonics by using the frequency-doubled output of the infrared signal from the OPG, instead of the sum-frequency signal generated in the KD\*P crystal. An alternative is to change the wavelength of the Ti:S laser to access any desirable tuning range.

Figure 3 presents an investigation of sum versus difference frequency mixing in different wavelength ranges, from xenon with high yields and low photon energies, via argon in the intermediate range, to neon with lower yields and high photon energies. The number of photons produced in each process is plotted as a function of the energy of the emitted radiation. There are several things to be learned from the figure. First, note that the two-colour signals exhibit the same characteristic features as the harmonics, namely a plateau and a cut-off, as also observed in the experiments involving a fundamental and its second or third harmonic (Eichmann *et al* 1995, Perry and Crane 1993). Since the plateau in neon can extend far beyond what we have observed in this experiment, we believe that the extension of the frequency mixing processes, and thereby the tunability, to the 150 eV range is a question of optimization of the conditions for generation of high-order harmonics. In the quasi-classical two-step interpretation of high-order harmonic generation (Corkum 1993, Schafer *et al* 1993) an electron is first released by tunnelling ionization, then accelerated by the electric field, before recombining with the nucleus and thereby emitting a harmonic photon. Thus, it is the maximum kinetic energy of the returning electron that determines



**Figure 3.** The number of photons as a function of the photon energy, for the different processes in xenon, argon and neon.  $(2N + 1)$  refers to the harmonic number  $2N + 1$  of the infrared, whereas  $(2N) + 1$  refers to the signal with frequency  $2N\omega_{\text{IR}} + \omega_{\text{OPG}}$ .

the cut-off. Therefore the two-colour signals are expected to exhibit a similar plateau and cut-off behaviour to one-colour harmonics, as is indeed observed in figure 3. Even the previously noted tendency, that the cut-off is less steep when the green field is present, might be interpreted as an additional acceleration of the electrons by the weak green field.

Second, note the change in the relative strengths of the  $+1$  and  $-1$  processes for increasing photon energies. In xenon, between 15 and 30 eV, the  $+1$  signals are clearly dominating, being an order of magnitude stronger than the  $-1$  signals, and the  $+2$  peaks

are almost as high as the  $-1$  peaks. In argon, between 20 and 40 eV, the two processes yield peaks of comparable strengths, the  $+1$  signals being stronger at low energies and the  $-1$  signals taking over at higher energies. Finally, in neon, above 40 eV, the  $-1$  signals are dominating the  $+1$  signals by about a factor of 5 and are even approaching the strength of the harmonic signals in the cut-off region. As mentioned previously,  $-2$  peaks are present, barely above the noise level, whereas no  $+2$  peaks can be detected.

At present, we have no explanation for this behaviour. Phase matching is predicted to be more efficient for difference frequency mixing schemes than for sum frequency mixing (Shkolnikov *et al* 1993, 1995, Bjorklund 1975). Indeed, in difference frequency mixing, the geometrical phase shifts owing to the two colours have opposite signs, and therefore partly compensate, leading to better phase matching. In contrast, in sum frequency processes, the geometrical phase shifts have the same sign and therefore add, leading to poorer phase matching. However, this difference between sum and difference frequency mixing is expected to *decrease* with the process order (simply, when the number of photons  $q$  for the first colour becomes large,  $q\phi_1 + \phi_2 \sim q\phi_1 \sim q\phi_1 - \phi_2$ ). This is at variance with the present experimental observations. The observed behaviour is therefore expected to originate in the single-atom dynamics. A theoretical analysis to interpret the experimental results is in progress.

In conclusion, we have studied sum and difference frequency mixing in the 15–70 eV range and have observed that the mixed signals behave as harmonic signals, concerning plateau, cut-off energy, and focusing conditions. We observe a marked change in the relative strength of the  $+1$  signals and the  $-1$  signals as the photon energy is increasing. Finally, we believe that the present tunability in the 7–70 eV range can be easily extended to cover the entire range from 7 to 150 eV.

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