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ANTI-STOKES RAMAN LASERS: A CONCEPT FOR THE GENERATION OF SHORT-WAVELENGTH COHERENT RADIATION

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Abstract

The anti-Stokes Raman process allows frequency up-conversion of coherent pulsed or cw radiation. For the process, a population inversion on a two-photon transition is needed. The principle of this technique, the present experimental situation and some developments will be described and discussed.

Introduction

The generation of coherent radiation at short wavelengths, in the vacuum-ultraviolet (vuv) or even soft x-ray (xuv) spectral range, is a difficult task and a still challenging problem in quantum optics. Principle difficulties thereby come from inherent frequency scaling laws of the laser. So, the pump power density in order to generate sufficient gain at a transition with frequency \( \nu \) scales proportional to \( \nu^4 \) or even \( \nu^5 \), for an inhomogeneous or a homogeneous lineshape respectively. For the realization of short-wavelength lasers, therefore, extremely powerful excitation processes are needed. Coupled with this are severe technological problems. A variety of concepts is being followed and investigated /1/2/. Among these, the anti-Stokes Raman laser technique seems to be of special interest, due to its great variety of possibilities.

Principle

The anti-Stokes Raman process takes place in a \( \lambda \)-type three level scheme as shown in Fig. 1. A necessary condition for operation of an anti-Stokes Raman laser (ASRL) is a selective excitation of the level 2 and the creation of a population inversion between the levels 2 and 1, where in general the level 2 has to be a metastable level. If this inversion is established, coherent pump radiation of frequency

\[
\Delta \nu
\]

\[
\nu_p
\]

\[
\nu_{AS}
\]

\[
\Delta E
\]

\[
1
\]

\[
3
\]

Fig. 1:

Anti-Stokes Raman laser scheme.
\( v \) can be converted (frequency up-shifted) into coherent radiation of frequency \( v_{AS} \) whereby a two-photon process between the levels 2 and 1 occurs. Basically, this process is a nonlinear (Raman-type), non-parametric process, however, at resonance \( (\Delta v = 0) \) also population is transferred from level 2 to level 3 and a population inversion between the levels 3 and 1 may be created. Therefore, at resonance this scheme may also be considered as an ordinary laser scheme, with a special two step excitation process, involving a metastable storage level. Thereby, the total excitation requirements may be reduced as compared to a direct one step excitation of the upper laser level.

The gain at the anti-Stokes frequency \( v_{AS} \) may be calculated by using the density matrix formalism \( /3/ \). In general, two gain contributions will exist, due to the Raman-type nonlinearity and due to an induced population inversion (normal laser gain), which however disappears at larger detunings \( \Delta v \). From investigations on Raman-type lasers it is well known, that even at resonance the Raman-gain contribution may be much larger than the gain contribution due to the induced inversion \( /3/ \). Thus, the considered scheme may give a higher gain than a comparable normal laser scheme. This can be of special importance for the operation of cw lasers at short wavelengths, where normally only a low inversion is possible.

For a detuning \( \Delta v \) large compared to the bandwidth of the involved transitions, only the Raman gain contribution remains. In this regime the gain \( \alpha \) is given by \( /4/ \):

\[
\alpha = K \frac{\Delta N \cdot I_p}{(\Delta v)^2}
\]

where \( \Delta N \) is the inversion density between the levels 2 and 1, \( I_p \) the pump intensity and \( K \) a constant which is proportional to the oscillator strength of the involved transitions and inversely proportional to the Raman linewidth. The above given formula is well suited for a calculation of the threshold pump intensity at a given detuning or of the tuning range at a given pump intensity.

**Features**

The anti-Stokes Raman laser process has the following main features:

1.) It is a non-parametric process and therefore requires no index matching.
2.) It allows tuning, by tuning the pump radiation.
3.) It allows large frequency shifts in a single step and is in principle suited for the vuv, the xuv or even the \( \gamma \)-ray spectral range.
4.) The process allows the conversion of pulsed and short-pulsed but also of cw laser radiation.
5.) The process allows high conversion efficiencies and large output energies as well.

**Requirements**

For the realization of an ASRL the following topics have to be considered and solved:

1.) An appropriate three level scheme with suitable resonances has to be found. For wavelengths down to about 50 nm, molecules and atoms may be used, for much shorter wavelengths ions have to be considered.
2.) A basic step is the production of the necessary population inversion between the metastable starting level and the final level. For this, a suitable selective excitation process of the meta-
In order to perform the conversion and obtain a powerful ASRL, a powerful pump radiation source in the range of the pump transition frequency has to be found.

**Inversion techniques**

Basically, all inversion techniques that are applied for normal lasers may also be used for the ASRL. For vuv and xuv ASRL the following techniques are mostly discussed and considered.

1.) Photodissociative production of the inversion.

A suitable molecule AB is photodissociated by suitable uv-radiation.

\[ AB + \text{hv} \rightarrow A^* + B \]

In this process, the species A (atom or molecule) is directly produced in the desired metastable state. All so far realized ASRL (see below) have been operated by this technique. This method can be highly selective and allows high inversion densities.

2.) Charge exchange reactions.

\[ A^* + B \rightarrow A^* + B^+ + \Delta E \]

This reaction is used in several metal-vapor lasers /5/ and can have a high selectivity and a large cross section (for \( \Delta E \approx 0 \)).

3.) Innershell excitation (ionization) of atoms by soft x-rays.

With this technique different primary lasers have already been realized /2,6/ and high densities of metastable atoms and ions have been produced /2,7/. Various anti-Stokes schemes have been proposed and are presently investigated /2,8/. High intensities of soft x-ray radiation can be produced efficiently, by laser induced plasmas (irradiation of suitable target materials with high intensity laser radiation) /9/.

Some examples of realized and possible ASRL using the above mentioned inversion techniques will be given in the following.

**History and present status**

The use of the anti-Stokes process for frequency up-conversion has first been proposed by Vinogradov et al. in 1972 /10/, but it lasted until 1982 before a first ASRL (with atomic Tl) had been realized by White et al./11/. At present, systems with atomic Tl/11,12/, I/13/, In/14/, Br/15/, Sn/16/, Pb/16/, Se/17,18/ and S/18/ have been realized and investigated. In all cases the necessary metastable inversion has been produced by photodissociation of molecules like TlI, NaBr, SnCl₂, COSe or COS. The mostly investigated system is atomic Tl. In the Tl system a conversion of 535 nm radiation (\( 6^2P_{3/2} \rightarrow 7^2S_{1/2} \)) into 377 nm radiation (\( 7^2S_{1/2} \rightarrow 6^2P_{1/2} \)) is possible, and conversion efficiencies up to 70%, output energies up to 5 mJ and tuning ranges of about \( \pm 5 \) nm (at 377 nm) have been measured /12/. For the vuv spectral range (\( \lambda \leq 200 \) nm) only systems with atomic Br, I, Se and S are suited. In case of Br and I, however, the energy shift is only 3400 cm\(^{-1}\) and 7600 cm\(^{-1}\) respectively, so that for these elements already short uv or vuv pump radiation is needed (for example 157 nm for Br/15/ and 206 nm for I/13/). At present, only the Se and S systems, with energy shifts up to 22000 cm\(^{-1}\), allow the conversion of uv radiation deep into the vuv. With these systems, the so far shortest anti-Stokes laser wavelengths of 146 nm (Se; pump wavelength 205 nm) and 148 nm (S; pump wavelength 219 nm) have been generated /17,18/. With the homologous oxygen system, a maximum energy shift of even 33000 cm\(^{-1}\) is possible, and anti-Stokes laser wavelengths down to 100 nm appear feasible (see Fig.2).
A high power vuv - ASRL with atomic Selenium

For all group VI elements (see Fig. 2) the interesting metastable level is the $^1S_0$ level. Possible anti-Stokes laser cycles either may end in the $^1D_2$ or the $^3F$ ground state. For selective population of the $^1S_0$ level, photodissociation of the molecules COSe, COS or N$_2$O may be used. The photodissociative reactions have been investigated by G. Black et al. /19/. In case of COSe, the optimum photodissociation wavelength is around 172 nm (quantum efficiency for $^1S_0$ production is close to 100%). In our experiments we used an ArF excimer laser (193 nm), which only gives a 30% efficiency for $^1S_0$, but allows high photodissociation fluences and the inversion of larger volumes. A setup for high power operation with a tunable narrowband ArF laser (oscillator-amplifier version) is shown in Fig. 3. The ArF laser is used for the photodissociation as well as for the production of the pump radiation. The pump radiation is generated by Raman conversion of the ArF laser in a H$_2$ or D$_2$ cell. In case of H$_2$, the 3rd Stokes component at 254 nm is exactly coincident with the $^1S_0$-$^3P_1$ pump transition of Se, and in case of D$_2$, the first Stokes component at 205 nm coincides with the $^1S_0$-$^3P_1$ pump transition of Se. Achieved data are summarized in Table I. As the above system contains only one primary laser system and two passive gas cells, easy and stable operation is possible. By further optimization output energies in the mJ-range appear feasible.

![Fig.2: Energy level schemes of Se, S and O](image)

Underlined: realized pump- and anti-Stokes wavelengths.

Fig. 3: Experimental setup for a high power Selenium ASRL.
Table I: Data and operation conditions of a high power Se-ASRL

Cell length 10 cm; typical OCSe pressure 0.3 mbar; Photodissociation pump energy 80 mJ.

Pump transition $^1S_0 - ^5s^3D_1 : 254$ nm $^1S_0 - ^5s^3P_1 : 205$ nm

ASRL transition $^5s^3D_1 - ^3P_1 : 167$ nm $^5s^3P_1 - ^3P_0 : 146$ nm

Pump energy 20 mJ 15 mJ
Output energy 100 µJ 350 µJ
Conversion efficiency 0.5% 2.3%
Peak power 50 kW 170 kW

Developments

An interesting candidate for an ASRL at 58 nm is the Helium atom (Fig. 4). For selective population of the metastable $2s^1S_0$ level, a charge exchange reaction between $He^+$-ions and alkali atoms may be used. Investigations of this reaction by Reynaud et al./20/ indicate, that for low energy $He^+$-ions (<1 keV) and for collisions with Rb and Cs atoms only the $2s^1S_0$ and $2s^3S_1$ metastable levels are populated. Especially the nearby $2p^1P_1$ level is not populated, which is important, as this level has a strong radiative decay into the $^1S_0$ ground state. The feasibility of this system is presently studied.

Many proposals exist to use inner shell ionization by soft x-rays for anti-Stokes Raman laser purposes/2,8,21/. Some possible schemes with atomic Li, Hg and Tl are shown in Fig. 5. Investigations of the Li system have been performed /22/ and are going on, our attempts concentrate on Hg and Tl. In case of Li and Hg, the anti-Stokes cycle ends in the ionic ground state. This ionic ground state is initially not populated by the soft x-rays, but will be rapidly populated by photoelectrons and plasma electrons. Short pulse plasma (soft x-ray) production will therefore be necessary and inversion is expected for a small time interval only. These problems may be avoided in case of Tl, where the final level lies well above the ionic ground state.

![Level scheme of Helium](image)
Fig. 5: Possible anti-Stokes laser schemes in atomic Li, Hg and Tl

**CW - ASRL**

Basically, the ASRL scheme is also suited for a conversion of cw radiation. However, a direct use of the pulsed schemes and inversion techniques will not be possible, and other schemes and techniques have to be found. Also inversion densities as high as for pulsed systems will not be possible (and necessary), and optical resonators have to be used. A direct consequence of this is, that metastable levels are no longer necessary, as amplified spontaneous emission processes will be negligible at lower inversion densities. This gives a larger freedom for possible systems. At present, the potential of existing cw laser materials, especially discharge pumped, for ASRL purposes is investigated. Three possible schemes (Fig. 6), using a metastable starting level, a cascade laser scheme or even the single photon inversion on normal laser transitions are considered. The cascade scheme is presently investigated at the He-Ne laser discharge, the hyper-Raman type (or two-step) up conversion scheme at the argon laser discharge. In this case the strong inversion at the $4p^2D_{5/2} - 4s^2P_{3/2}$ - transition, which drives the 488 nm laser will

![Diagram of energy levels and transitions](image-url)
be applied (see Fig.7). By use of two Rhodamin 6G dye lasers a generation of radiation at 191.3 nm appears feasible. A cw ASRL operating between the metastable 3d^2F - levels and the 4s^2P_{3/2}-level of Ar with the intermediate 4p^2S_{1/2}-level (see Fig.7), converting 648 nm radiation into 437.5 nm radiation has already been realized. In first experiments an output power of 130 mW, a conversion efficiency of up to 20 % and a tuning range of ±12 GHz has been realized. In a ring resonator also unidirectional oscillation, as expected, is observed. By using higher lying metastable levels (for example 3d^2P) also direct anti-Stokes schemes with much shorter wavelengths will be possible.

Fig.7: Level scheme of the argon ion with realized (--) and further possible (---) up-conversion schemes.

Conclusions
The ASRL technique offers a variety of possibilities for the generation of pulsed as well as cw coherent radiation at short wavelengths. The special potential of this technique seems to lie in the capability to generate powerful sources with a high conversion efficiency. The tuning aspects, however, are not so pronounced and tuning will in general be limited to small regions around resonances. Anti-Stokes Raman lasers may therefore be considered as special optically pumped laser sources.

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