Vol 18 No 10, October 2009
1674-1056/2009/18(10)/4308–06

Chinese Physics B

© 2009 Chin. Phys. Soc.
and IOP Publishing Ltd

Efficient collinear frequency tripling of femtosecond laser with compensation of group velocity delay*

Wang Yan-Ling(王燕玲), Zhou Xu-Gui(周绪桂),
Wu Hong(吴 洪), and Ding Liang-En(丁良恩)†

State Key Laboratory of Precision Spectroscopy and Department of Physics, East China Normal University, Shanghai 200062, *China*

(Received 16 December 2008; revised manuscript received 26 February 2009)

This paper demonstrates an approach that negative uniaxial crystal has a relative anomalous dispersion effect which can compensate group velocity delay, and applies this approach to nonlinear frequency conversion of an ultrafast laser field. High efficiency of the third harmonic generation is experimentally fulfilled by adopting a collinear configuration of doubing-compensation-tripling system. Through finely adjusting the incident angle and optical axis direction of the compensation plate, it obtains ultraviolet (UV) output energy of 0.32 mJ centered at 270 nm with spectral bandwidth of 2 nm when input beam at 800 nm was 70 fs pulse duration and 6 mJ pulse energy which was extracted from Ti:sapphire laser system by a diaphragm, corresponding to an 800-to-270 nm conversion efficiency of 5.3% and a factor-of-1.6 improvement in the third harmonic generation of UV band in comparison with a general conventional configuration. Furthermore, when the full energy of 18 mJ from a Ti:sapphire laser system was used and optimized, the UV emission could reach 0.83 mJ.

Keywords: nonlinear frequency conversion, ultrafast laser, sum-frequency
PACC: 4265K

1. Introduction

With the development of high intensity ultrafast laser technology, intense ultraviolet (UV) output generated by nonlinear frequency conversion has become a centre of focus in recent years.[1−16] The most widely used method for the third harmonic generation is based on the second-order nonlinearity of crystal. In the year 2000, T. S. Kojima *et al* obtained UV power of 20.5 W with a repetition rate of 10 kHz by the use of a high-power all-solid-state green laser and a high-quality $CsLiB_6O_{10}$ crystal;[2] in the year 2003, H. Kitano *et al* demonstrated 3.0 W, 355 nm UV at 31 kHz with 30% conversion efficiency produced in CsB_3O_5 crystals grown by the top-seeded solution growth technique;[15] in the year 2007, Ebrahim-Zadeh and co-workers achieved 0.216 mJ in 29 ps duration at 25 Hz repetition rate UV output centered at 355 nm with overall conversion efficiency of up to 50%.[16] All these illustrate that the considerable nonlinear transformation has been already achieved for a long pulse duration of ns or ps. However, as for femtosecond pulses, it cannot obtain the high conversion efficiency of the third harmonic in collinear structure which is mostly limited by the disadvantages of temporal separation of coupled waves due to the large difference of group velocity, pulse duration broaden by group velocity dispersion and less nonlinear coefficient of type-II phase matching.

This paper demonstrates an approach that negative uniaxial crystal has a relative anomalous dispersion effect which can compensate group velocity delay, and this approach is applied to the nonlinear frequency conversion of ultrafast laser field. The high efficiency of the third harmonic generation (THG) is experimentally fulfilled by adopting a collinear configuration of doubing-compensation-tripling system. The compensation plate can reduce time delay, weaken group velocity dispersion effect on pulse duration, and influence phase-matching type so as to increase the third harmonic conversion efficiency. We achieved the single pulse energy of 0.32 mJ centered at 270 nm with a full width at half maximum (FWHM) of 2 nm under the condition that input energy extracted from the Ti:sapphire laser system by a diaphragm was 6 mJ and a spot diameter was 6 mm, attaining an 800-to-270 nm conversion efficiency of 5.3% and 1.6 times improve-

*Project supported by the National Basic Research Program of China (Grant No 2006CB0806001), the Program for Changjiang and Innovative Research Team in University, Shanghai Leading Academic Discipline Project (Grant No B408).
†Corresponding author. E-mail: leding@phy.ecnu.edu.cn

ment of the conversion efficiency in comparison with a conventional approach.

2. Principle

In the conventional way of the third harmonic generation, an input beam of central frequency of ω firstly generates the second harmonic in frequency doubler and then generates the third harmonic in tripler by sum-frequency between the second harmonic and residual input beam. After frequency doubling, the fundamental and second harmonic are polarized perpendicularly to each other, so sum-frequency process belongs to type-II phase matching. Nevertheless, as for femtosecond pulses, it is hard to attain the third harmonic translation efficiently by only using the above general collinear approach with a single frequency doubler and tripler. This is mostly attribute to the very short pulse duration which can make the temporal separation of coupled waves even longer than the pulse duration itself, and less type-II nonlinear coefficient etc. To overcome this shortage, the principle of the third harmonic generation with a compensation plate is shown in Fig.1, in which negative uniaxial crystal BBO is employed as this compensation plate, filling the role of compensating group velocity delay.

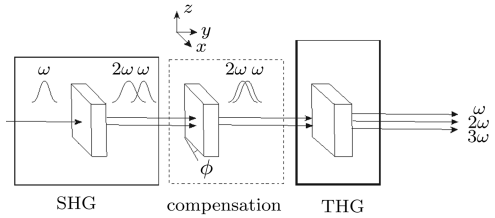


Fig.1. Compensation principle scheme of the third harmonic generation.

2.1. Compensation of group velocity delay using BBO crystal

While propagating through crystal BBO, an ordinary fundamental wave (o-wave) at 800 nm can generate extraordinary second harmonic (e-wave) at 400 nm. However, due to the large group velocity difference of femtosecond pulses, these two waves will separate quickly in time domain. When the fundamental wave is normally injected and BBO is cut with a type-I phase-matching angle of 29.2°, the time delay between 800 nm and 400 nm waves can be worked out: $\Delta u^{-1} = u_{400}^{-1} - u_{800}^{-1} = 194$ fs/mm. Further more, the third harmonic generation in sum-frequency crystal also has similar problem such as the time delay between 270 nm and 400 nm waves is: $\Delta u^{-1} = u_{267}^{-1} - u_{400}^{-1} = 390$ fs/mm. It can be seen that more crystal thickness will introduce more time delay. Since the third harmonic is generated only by the overlap of input pulses, we have to achieve the synchronization of fundamental wave and the second harmonic first, which would prevent significant decrease of conversion efficiency. In addition, the more the second harmonic is generated in the frequency doubling process through the crystal, more pulse duration of second harmonic will be broadened than fundamental wave. Pulse duration broadening can decrease peak intensity which goes against nonlinear effect, also leading to conversion efficiency and spot quality being affected. So the choice of a thinner crystal and realization of temporal overlap as much as possible become necessary, and we employed a thickness of 0.2 mm BBO crystal in our experiment, corresponding to time delay of 39 fs.

According to index ellipsoid of negative uniaxial crystal, the time delay between the ordinary and extraordinary wave is dependent on different optical paths and the refractive index of an extraordinary wave which is changed with the angle of incidence. Reference [17] describes the propagation solved by Maxwell's equations in the case of slanting incidence, and it can be further simplified in our application. Figure 2 presents the propagation under the condition of slanting incidence where ϕ is incident angle and θ is phase-matching angle. Obviously, depending on the angle between incident direction and orientation of optical axis, the extraordinary wave can propagate in a direction closer to, or further away from the normal surface than the ordinary wave, so the different incident angle leads to variation of optical paths. Moreover, according to refractive index formula for extraordinary wave: $n_{e(\theta')} = \frac{n_e n_o}{\sqrt{n_o^2 \sin^2(\theta') + n_e^2 \cos^2(\theta')}}$, where θ' is the angle between input direction and optical axis. It can be seen that different incident angle also leads to variation of extraordinary refractive index. According to Ref.[18]: $\tan\alpha = [Q_e \sin\theta \sin\phi - (\varepsilon_0 - Q_e^2)\cos\theta]/(Q_o^2 \sin\theta - Q_e \sin\theta \sin\phi)$, $\tan\alpha_k = \sin\phi/Q_e$, where α_k is k-vector direction for extraordinary wave, and $Q_o^2 = \varepsilon_o - \sin^2\varphi$, $Q_e = [D - (\Delta\varepsilon/2)\sin 2\theta \sin\varphi]/(\varepsilon_o + \Delta\varepsilon\cos^2\theta)$, $D = [\varepsilon_o \varepsilon_e(\varepsilon_o + \Delta\varepsilon\cos^2\theta - \sin^2\varphi)]^{1/2}$, $\varepsilon = n^2$. Here $\theta' = \alpha_k + \theta$, combining with the law of refraction: $\sin\beta = \sin\varphi/n_o$ and group velocity reciprocal for-

mula: $u^{-1} = n/c - (\lambda/c)(\mathrm{d}n/\mathrm{d}\lambda)$, the time delay between two waves can be calculated by $\Delta t = t_{\mathrm{o}} - t_{\mathrm{e}} = OA/u_g^{\mathrm{o}} + AC/c - OB_{\mathrm{eff}}/u_g^{\mathrm{e}}$, where OA and AC are optical paths of ordinary wave, while OB_{eff} is optical path of extraordinary wave.

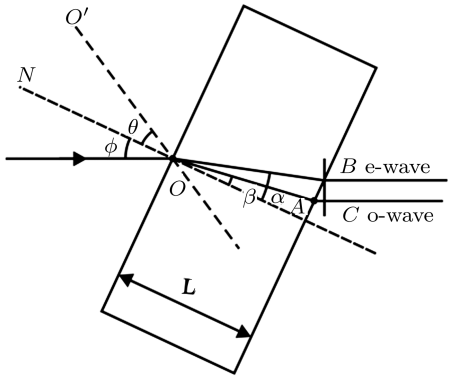


Fig.2. Propagation with incident angle ϕ.

We calculated the relationship between time delay and incident angle ϕ in the case of BBO crystal with 0.2 mm thickness and type-I phase-matching angle of 29.2° (see Fig.3).

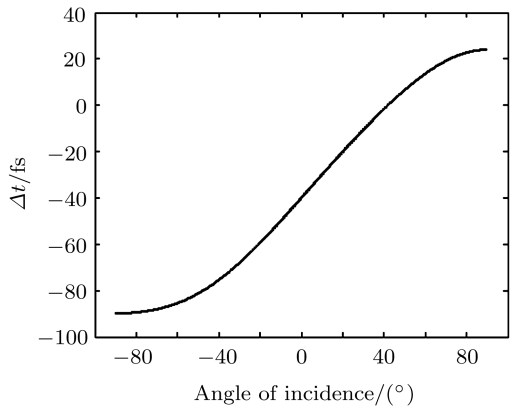


Fig.3. Dependence of time delay on incident angle ϕ (0.2 mm BBO, 29.2°).

X-axis represents an incident angle ϕ, y-axis represents time delay Δt between 800 nm and 400 nm. When $\Delta t < 0$, it means 400 nm lags behind 800 nm, whereas 800 nm lags behind 400 nm. It can be seen that a time delay is about 39 fs when normal incidence $\phi = 0°$, and this compensation of time delay is realized when incident angle increases to nearly 40°. The extent of compensation will be different with variation of incident angle. Therefore, the time delay between these two pulses can be corrected with a tunable velocity compensation plate of nonlinear crystal BBO.

2.2. Correcting the polarization

In a conventional approach, the polarization of the fundamental wave and second harmonic out from the frequency doubler are perpendicular to each other which means that the sum-frequency process has to be type-II phase matching which has a less effective nonlinear coefficient than type I.

In fact, a birefringent crystal BBO also fills the role of rotating polarization by turning it into the propagation direction which leads to the variation of principal section. Corresponding to a different principal section, the polarization will be changed. If $p_\perp$ and $p_\parallel$ respectively stand for perpendicular and parallel polarization to the principal section, then according to Fig.4, the polarization will be given by

$$p_\perp = p_{800}\cos\gamma + p_{400}\cos\left(\frac{\pi}{2} - \gamma\right),$$

$$p_\parallel = p_{800}\sin\gamma + p_{400}\sin\left(\frac{\pi}{2} - \gamma\right).$$

where γ is the angle between projection of turning angle and original optical axis of optimum phase-matching in crystal surface, y axis is the propagation direction, θ is the angle between wave propagation direction and optical axis. The adjustment of optical axis direction of compensation plate can make variation in the polarization, and the nonlinear frequency conversion efficiency is also affected correspondingly. Actually, while the rotating compensation plate about the propagation direction, the two waves of 800 nm and 400 nm may have a little spatial offset. However, it can be ignored due to a comparatively larger spot diameter.

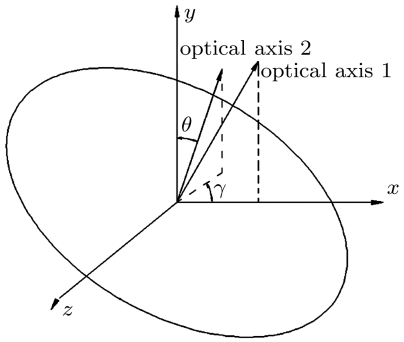


Fig.4. Relation of different optical axis directions.

3. Experiments

The schematic diagram of experimental setup is presented in Fig.5. A Ti:sapphire (TSA) laser system (TSA-25, Spectra-Physics Inc., USA) which is based

on chirped-pulse amplification, is employed as the fundamental light source operated at 10 Hz of frequency repetition with spectrum centered at 800 nm and pulse duration of 70 fs. The energy of single pulse extracted by a diaphragm here is about 6 mJ, corresponding to a spot diameter of 6 mm. BBO(1) and BBO(3) are frequency doubler and tripler respectively, while BBO(2) serves as a compensation plate.

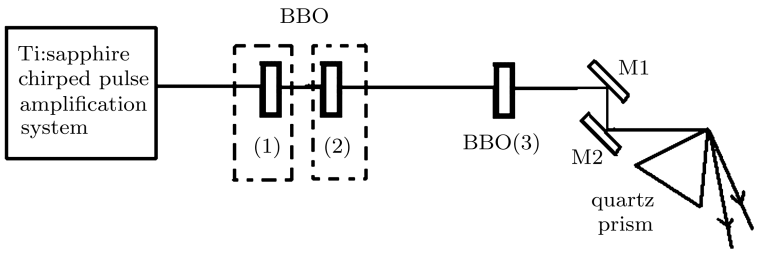


Fig.5. Experimental setup. BBO(1)(2):type-I phase-matching β–BBO cutting at 29.2°, 0.2 mm; BBO(3): type-I phase-matching β–BBO cutting at 44.3°, 0.2 mm; M1, M2: 45° high reflective mirrors at 267 nm with broadband; BBO(1)(2) are on individual translation stages independently.

In a conventional way of UV generation, a fundamental wave at 800 nm first produces the second harmonic at 400 nm in crystal BBO(1) and then generates the third harmonic in BBO(3) by sum-frequency between residual fundamental and generated second harmonic. Both a thickness of 0.2 mm, while cutting angles are individually 29.2° and 44.3°. The generated UV wave by sum-frequency is finally injected into the quartz prism with the Brewster angle to separate various wavelengths by dispersion. Through fine regulation, we obtained pulse energy of 1.5 mJ at 400 nm, achieving an 800-to-400 nm conversion efficiency of 25%, and then fulfilled maximum UV emission of 0.2 mJ from BBO(3). The spectrum width is 2 nm (see Fig.6(a)) which can support Gaussian pulses

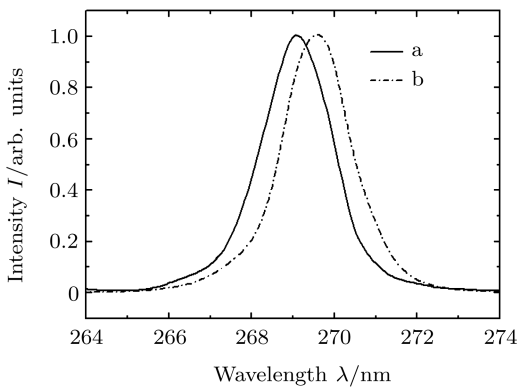


Fig.6. UV spectrum.

duration of 52 fs under Fourier transform limit condition. Due to depolarization of crystals, the polarization relationship between 400 nm and 800 nm or 270 nm measured was approximately perpendicular, and the direction between 800 nm and 270 nm was nearly parallel which demonstrated the sum-frequency process should belong to a type-II interaction.

In order to compensate the group velocity delay, we added another identical crystal BBO(2) after BBO(1), the separation between these two crystals was only 7 mm so as to decrease phase shift caused by air. Rotating crystal BBO(2) about the injecting direction of a fundamental wave could lead to the variation of the optical axis, and tilting the crystal surface a little could induce the change of incident angle. All of these brought about the different optimum situation of BBO(3) to obtain the most intensive energy of UV output. In this experiment, we achieved a UV pulse energy of 0.32 mJ in the case of the second harmonic pulse energy of 1.6 mJ, and 26% conversion efficiency produced after these two crystals. The generated UV radiation was 1.6 times of the energy of conventional method. Obviously, the significant increase of frequency tripling efficiency was not due to frequency doubling growth which varied very little here, it should be due to compensation of group velocity delay. Figure 6(b) shows that the spectrum width is also 2 nm and the center of the third harmonic spectrum is red-shifted about 0.5 nm in comparison with Fig.6(a), which means that the optimum wavelength of phase matching has a movement in sum-frequency process. The polarization between 800 nm and 270 nm almost became vertical, the direction of

800 nm rotated by 68° and 400 nm could not be measured, meaning that the sum-frequency belonged to type I basically.

To show the compensation role of BBO(2) adequately, we used a cross-correlation setup (see Fig.7) to measure the delay between 400 nm and residual 800 nm after frequency doubling and their respective temporal width. The TSA output was firstly split into

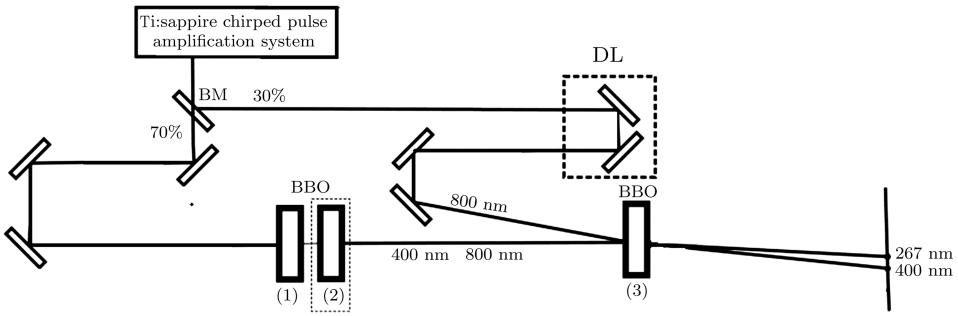


Fig.7. Cross-correlation setup.

two beams by 3:7 beam splitter (BM). The weak beam of 800 nm serves as a reference beam through a delay line mixed frequency with the second harmonic (400 nm) and residual fundamental wave (800 nm) simultaneously which both realized in BBO(3), leading to the output at 270 nm and 400 nm respectively, in which 270 nm wave out from BBO(3) actually reflected the second harmonic of 400 nm, while 400 nm wave from BBO(3) reflected the residual 800 nm after BBO(1). By analyzing the temporal width and delay between 270 nm and 400 nm, the distinguished information of 400 nm and 800 nm pulses in process of frequency doubling with single crystal or double crystals could be seen in Fig.8, solid lines BBO 1–400 –■– and 1–800 —□— reflect the cross-correlation temporal width of the second harmonic and residual fundamental wave produced only by BBO(1), dash lines BBO 12–400 - -▲- - and 12–800 - -△- - reflect those results of BBO(1) and BBO(2). For frequency doubling with one crystal, the cross-correlation temporal width of 400 nm (570 fs) which lags behind about 100 fs is much broader than that of 800 nm (440 fs). While as for cascading doubling crystals, 400 nm wave lags behind 800 nm wave only 30 fs, and both their cross-correlation widths decrease to about 350 fs. All these illustrate the compensation role of group velocity delay which makes great contribution to the third harmonic generation. Actually, we could not fulfill the compensation of temporal separation completely. Nevertheless, great improvement of temporal overlap increasing by 2/3 through this approach could be realized.

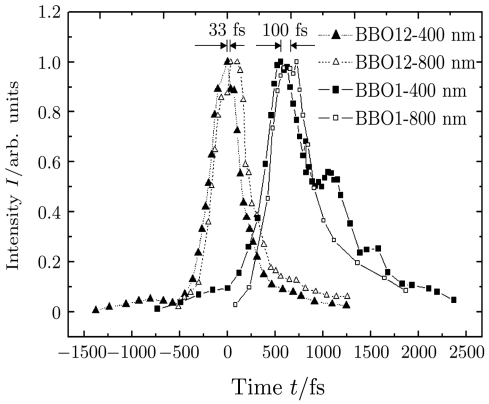


Fig.8. The time delay of cross-correlation measurement. symbols ■ and □: temporal features of the second harmonic and residual fundamental wave propagated in a single BBO; symbols ▲ and △: temporal features of the second harmonic and residual fundamental wave propagated in double BBOs.

Furthermore, when total energy from TSA system of 18 mJ was injected into BBOs, the single pulse energy of 270 nm could reach 0.83 mJ, which is a comparatively high energy source in this wave-band for femtosecond laser at present.

4. Conclusions

In conclusion, we have demonstrated the rationality of the compensation of group velocity delay by adding a compensation plate BBO(2). This method can decrease the time delay of interactional pulses, lessen pulse duration broadening and influence phase-matching type, leading to increase the third harmonic generation. In the experiment, we obtained UV pulses with single pulse energy of 0.32 mJ, 800-to-270 nm conversion efficiency of 5.3% and spectrum width of 2 nm in the case of input energy of 6 mJ at 800 nm. When the total energy of 18 mJ out from TSA system was injected into BBOs, a single pulse energy of 270 nm could reach 0.83 mJ. Meanwhile, we also employed an approach of two-photon fluorescence to measure generated UV pulse duration which was approximately 200 fs.

References

[1] Liu Y Q, Zhang J, Liang W X and Wang Z H 2005 *Acta Phys. Sin.* **54** 1593 (in Chinese)

[2] Tetsuo K, Susumu K, Shuichi F, Koji Y, Kenji Y, Yusuke M, Takatomo S, Mitsuhiro T and Yukikatsu O 2000 *Opt. Lett.* **25** 58

[3] Gao L L and Tan H M 2003 *Optoelectronic Technology* **23** 109 (in Chinese)

[4] Fan X W, Wang Y, Peng Q Q, Liu J and He J L 2005 *Laser and Infrared* **35** 331 (in Chinese)

[5] Wang Y, Fan X W, Peng Q Q, Liu J and He J L 2005 *Optoelectronics Laser* **16** 551 (in Chinese)

[6] Tan C Q, Zheng Q, Xue Q H, Bu Y Q, Jia F Q and Qian L S 2005 *Laser and Infrared* **35** 490 (in Chinese)

[7] Tan C Q, Xue Q H, Jia F Q, Qi Y, Zheng Q and Qian L S 2005 *Acta Photonica Sinica* **34** 1289 (in Chinese)

[8] Wu Y C, Chang F, Fu P Z, Chen C T, Wang G L, Geng A C, Bo Y, Cui D F and Xu Z Y 2005 *Chin. Phys. Lett.* **22** 1426

[9] Zhang S J, Dong S M, Li F Q, Zhai Z J, Xu X G, Wang J Y, Wang Q P, Zhang F J, Zhang X Y and Li P 2003 *Chinese Journal of Lasers* **30** 1087 (in Chinese)

[10] Ma J, Zhang R B, Liu B, Zhu C, Chai L, Zhang W L, Zhang Z G and Wang Q Y 2005 *Acta Phys. Sin.* **54** 3675 (in Chinese)

[11] Zhao S L, Zhu B Q, Zhan T Y, Cai X J, Liu R H, Yang L, Zhang Z X and Bi J J 2006 *Acta Phys. Sin.* **55** 4170 (in Chinese)

[12] Mizuuchi K, Morikawa A, Sugita T, Yamamoto K, Pavel N and Taira T 2004 *Appl. Phys. Lett.* **85** 3959.

[13] Yang Y S, Zheng W G, Han W, Che Y L, Tan J C, Xiang Y and Jia H T 2007 *Acta Phys. Sin.* **56** 6468 (in Chinese)

[14] Feng Y, Bi Y, Zhang H B, Yao A Y, Wang G L, Lin X C, Wang J S, Xu Z Y, Zhu Y and Chen C T 2002 *Chinese Journal of Lasers* **29** 766 (in Chinese)

[15] Kitano H, Matsui T, Sato K, Ushiyama N, Yoshimura M, Mori Y and Sasaki T 2003 *Opt. Lett.* **28** 263

[16] Ebrahim-Zadeh M 2007 *IEEE Journal on Selected Topics in Quantum Electronics* **13** 679

[17] Lekner J J 1991 *J. Phys: Condensed Matter* **3** 6121

[18] Henrik Enqvist 2004 *Lund Reports on Atomic Physics* p10