Appl. Phys. B (2004)
DOI: 10.1007/s00340-004-1428-3

Applied Physics B
Lasers and Optics

Y. UESU[1]
K. YASUKAWA[1]
N. SAITO[1]
S. ODOULOV[2]
K. SHCHERBIN[2,✉]
A.I. RYSKIN[3]
A.S. SHCHEULIN

Backward-wave four-wave mixing and coherent oscillation in CdF_2:Ga, Y

[1] Physics Department, Waseda University, Tokyo, Japan
[2] Institute of Physics, National Academy of Sciences, 03650, Kiev-39, Ukraine
[3] S.I. Vavilov State Optical Institute, 199034 St. Petersburg, Russia

Received: 29 August 2003/Revised version: 18 January 2004

ABSTRACT Cadmium fluoride doped with Ga and Y is shown to be a suitable medium for backward-wave four-wave mixing with a grating decay time of a few seconds at room temperature. A 3-mm-thick sample ensures amplified phase-conjugate reflectivity and can therefore be used for building a coherent oscillator with a conventional mirror.

PACS 42.65.Hw; 42.70.Mp; 42.70.Nq

1 Introduction

The semiconductors CdF_2:In, CdF_2:Ga, and CdF_2:Ga,Y possess at ambient temperature a strong third-order nonlinearity caused by reversible optical transformations of the deep DX centers [1–3]. The light-induced refractive-index variation is attributed to the conversion of a DX center from its ground (two-electron deep) Ga^{1+} state into the metastable (shallow donor) state, in which an electron is weakly bound at the hydrogenic orbital centered at the Ga^{3+} ion. The photogenerated weakly bound electrons affect the crystal plasma frequency in a similar way as free carriers do [4, 5] and therefore the high-frequency dielectric constant becomes modified. The refractive-index change is negative (which was confirmed by z-scan measurements) and increases linearly with the light intensity [3].

It has been shown recently that at room temperature CdF_2:Ga,Y possesses a high nonlinear susceptibility $\chi^{(3)} \approx 1.4 \times 10^{-10}\,\mathrm{m}^2/\mathrm{V}^2$ and a relatively short response time of the order of a few seconds [2, 3]. These features ensure the recording of dynamic phase gratings with a diffraction efficiency approaching 50% and make CdF_2:Ga, Y a potential candidate for different wave-mixing applications. The purpose of this paper, in particular, is to study backward-wave four-wave mixing (BWFWM), which is the underlying process for optical phase conjugation and also for various kinds of frequency-degenerate coherent optical oscillators.

2 BWFWM in media with third-order nonlinearity

If a crystal with third-order nonlinearity is pumped with two counterpropagating coherent light waves 1 and 2 (as shown in Fig. 1a) the signal wave 4 sent to the illuminated region gives rise to the appearance of the backpropagating wave 3. For a pair of the pump waves with mutually conjugate wavefronts the generated wave 3 is a phase-conjugate replica of the signal wave 4. The ratio of the intensities of these two waves is called the phase-conjugate reflectivity, $R_{pc} = I_3/I_4(0)$. For nonlinear media where the reflection gratings are recorded with the same efficiency as transmission gratings the phase-conjugate reflectivity in the undepleted pump approximation is given by [6, 7]

$$R_{pc} = \frac{\sin^2(\gamma\ell)}{\cosh^2\left(\frac{\ln r}{2}\right) - \sin^2(\gamma\ell)}, \tag{1}$$

which is valid within the plane-wave approximation for all interacting waves and undepleted pump waves. Here $\gamma\ell = 2\pi\Delta n\ell/\lambda$ is the coupling strength, Δn is the refractive-index change, ℓ is the interaction length inside the sample, λ is the light wavelength, and r is the pump intensity ratio, $r = I_2/I_1$.

In the case that only one of the two index gratings is recorded, either transmission or reflection, the argument of the trigonometric functions of (1) becomes equal to $\gamma\ell/2$ [6, 8]. In both cases (with both types of grating recorded and with only one of the two) for a certain thickness of the nonlinear crystal λ the phase-conjugate reflectivity R_{pc} becomes larger than unity, i.e. the intensity of the phase-conjugate wave is larger than the intensity of the signal.

A phase-conjugate mirror with $R_{pc} \geq 1$ can form a coherent oscillator together with a conventional high-reflective mirror (see Fig. 1b). Self-oscillation develops if the threshold condition of oscillation is met:

$$RR_{pc} = 1. \tag{2}$$

Here R is the reflectivity of the conventional mirror.

The oscillation in a semilinear cavity has already been achieved in media possessing nonlocal response, in which case the index grating is $\pi/2$-shifted with respect to the interference pattern (photorefractive crystals with diffusion-driven charge transport or with charge hopping [9–11]). It was demonstrated also for media with various local optical nonlinearities (for example, with the Kerr nonlinearity in CS_2 [12], the resonant nonlinearity in gases [13], the thermal nonlinear-

---

✉ Fax: +380-44/265-2359, E-mail: kshcherb@iop.kiev.ua

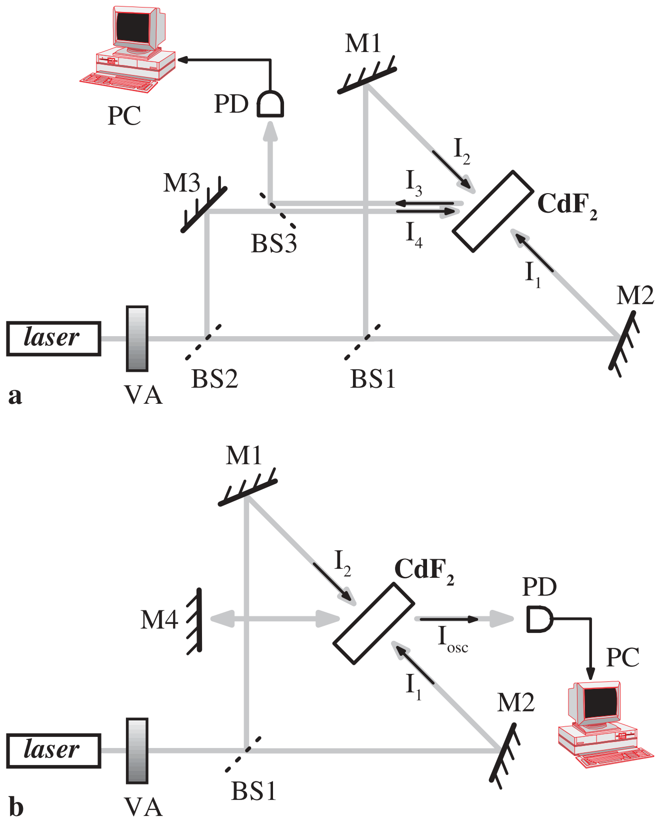

FIGURE 1 Experimental set-up for study of the backward-wave four-wave mixing (**a**) and oscillation in a semilinear cavity (**b**) in CdF$_2$:Ga, Y; BS – beam splitters, M – mirrors, VA – variable attenuator, PD – photodetector, PC – data-acquisition system controlled by a personal computer

ity in liquid crystals [14], and the free-carrier nonlinearity in semiconductors [15]).

The oscillator with one phase-conjugate mirror has the ability to compensate for intracavity phase distortions. This may be of practical importance for hybrid systems with the additional laser amplifiers inside the cavity with high gain but moderate optical quality [8, 16].

3 Experimental technique

The CdF$_2$ single crystals are grown by the Stockbarger–Bridgeman technique. To get a high density of the active Ga centers up to $1.7 \times 10^{18}\,\text{cm}^3$ the co-doping with Y was used; the co-doping also improves the crystal optical quality [17]. After being heated in a reducing atmosphere of K and Cd vapors the samples became photosensitive. They have a slightly greenish color with the absorption coefficient $\alpha \approx 0.7\,\text{cm}^{-1}$ and $n \approx 1.6$ at $\lambda = 0.53\,\mu\text{m}$.

The sample used in the present paper has two optically finished faces with spacing 3 mm between them. Two light beams from the cw frequency-doubled Nd^{3+}:YAG laser (single-mode, single-frequency, output power up to 100 mW) enter the sample as shown in Fig. 1a. The illuminated area on the sample input face has a diameter about 3 mm ($1/e^2$ intensity level in Gaussian distribution). The signal beam 4 impinges upon the crystal at an angle $\theta \approx 3.3°$ to the beam 2. The intensity of the beam 3 is 10 times smaller than the total pump intensity. All beams are identically polarized perpendicularly to the plane of incidence. A variable attenuator VA placed between the laser and the beam splitter is used to control the recording light intensity. The calibrated filters are used to change the intensities of both recording beams. Together with the attenuator these filters allow changing the pump intensity ratio keeping the total intensity constant.

The coherence length of the pump laser is much larger than all path differences in the present experiment; thus, all types of gratings are recorded in the sample, reflection type as well as transmission type.

4 Experimental results and discussion

The phase-conjugate reflectivity is measured at first as a function of the intensity of pump waves (Fig. 2a). At small intensities R_{pc} increases roughly as I^2. This is in agreement with our expectations for a medium with third-order nonlinearity. The phase-conjugate reflectivity saturates with intensity at $I > 20\,\text{mW/cm}^2$ because of the crystal heating by the recording beams with relatively high intensity [3].

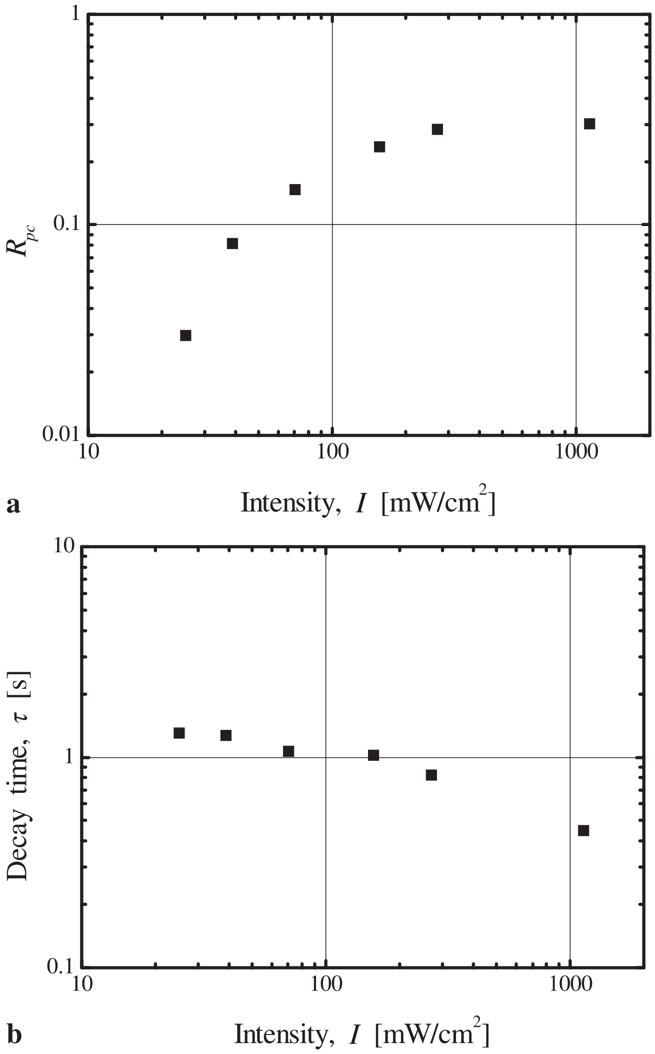

FIGURE 2 Phase-conjugate reflectivity (**a**) and decay time of the conjugated wave (**b**) as a function of the total light intensity, measured at angle $\theta \approx 3.3°$ and intensity ratio of the pump beams $r \approx 1$

In addition, the decay of the phase-conjugate wave is studied when the signal wave is stopped in the presence of the pump beams. As is seen from Fig. 2b, the decay time of the phase-conjugate wave is decreasing only by a factor of three within the whole range (nearly two orders of magnitude) of intensity. This decrease is similar to that observed in two-beam-coupling experiments; it is also related to the sample heating [3].

Figure 3 shows the pump-ratio dependence of the phase-conjugate reflectivity. A typical bell-shaped dependence is obvious, which is in agreement with the theoretical prediction [6]. From the fit of (1) to experimental data the coupling strength can be extracted: $\gamma\ell = 2\pi\Delta n\ell/\lambda \approx 0.55$. This value is however obviously underestimated, as will become clear from further experiments with coherent oscillation. We will discuss this point below, after a description of the experimental results.

With a conventional flat mirror with $R \approx 1$ placed at a distance of 5 cm from the sample in such a way that the normal to its surface makes an angle of about 3° with the direction of the pump waves, coherent oscillation has been achieved. However, the oscillation is only slightly above the threshold when the input sample face is aligned nearly normal to the pump and generated beams. In this case the oscillation beam is very weak and its intensity is unstable because of the mechanical instabilities: the oscillation beam appears and disappears chaotically. To go more above the threshold of the oscillation we tilt the sample to the angle of about 50° with respect to the oscillation beam and use horizontal polarization of the light. In this case the interaction length increases only slightly, but what is more important the Fresnel losses of the oscillating beam are reduced considerably as the angle of incidence approaches the Brewster angle (about 58° for CdF_2).

Figure 4 shows a temporal development of the oscillation wave intensity, which is characteristic for all coherent oscillators (see e.g. [9]). For a relatively long time after the beginning of the crystal exposure with two pump waves the measured signal is very small; starting from a certain delay time it increases and reaches its steady-state value. The well-pronounced intensity fluctuations in saturation should be attributed to poor mechanical stability of the optical table.

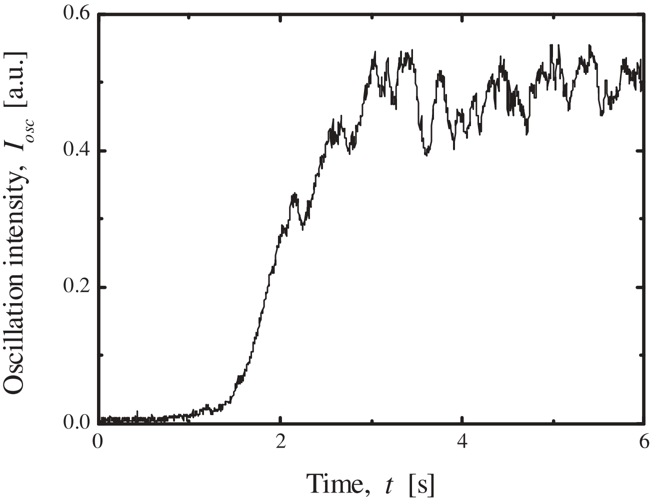

FIGURE 4 Temporal dynamics of the oscillation intensity, measured at angle $\theta \approx 3.3°$, total intensity $I \approx 200\,\mathrm{mW/cm^2}$, and intensity ratio of the pump beams $r \approx 1$

The onset of oscillation in a semilinear cavity proves unambiguously that the phase-conjugate reflectivity is at least higher than unity, i.e. amplified reflection is reached. In fact, the phase-conjugate reflectivity is somewhat larger because the oscillation is observed with a neutral density filter with the transmittance $T \approx 0.7$ placed inside the cavity. Neglecting all other cavity losses and taking into account that the oscillating beam passes through the filter twice, we can estimate R_{pc} as the effective reflectivity of a conventional mirror with the filter $R = T^2$, i.e. $R_{\mathrm{pc}} \geq 2$.

The independent confirmation that $R_{\mathrm{pc}} > 1$ is obtained from the pump-ratio dependence of the oscillation beam intensity (Fig. 5). The oscillation occurs within a certain range of r with the threshold pump ratios $r_{\mathrm{th1}} \approx 0.4$ and $r_{\mathrm{th2}} \approx 3$, at which the oscillation emerges from optical noise and can be detected with confidence. Taking $R = 1$ in the threshold condition of oscillation (2), we get from (1) the sample coupling

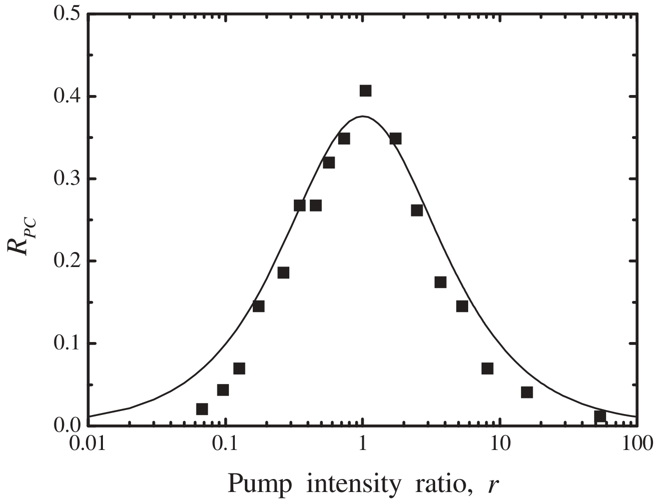

FIGURE 3 Phase-conjugate reflectivity (*squares*) as a function of pump intensity ratio, measured at angle $\theta \approx 3.3°$ and total intensity $I \approx 270\,\mathrm{mW/cm^2}$. *Solid line* represents best fit of (1) to experimental data

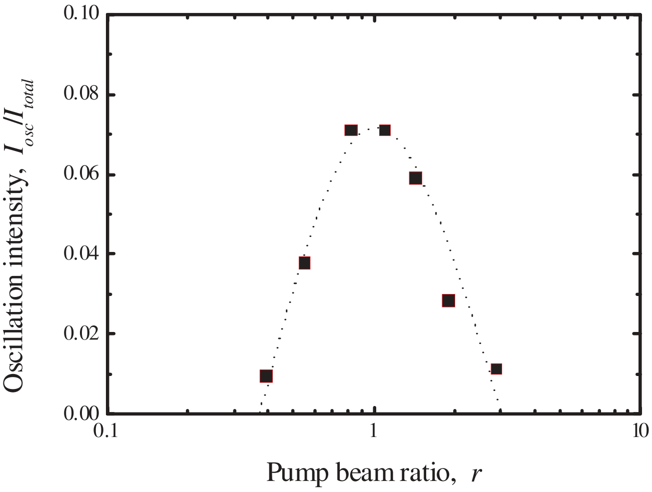

FIGURE 5 Oscillation intensity dependence on pump intensity ratio, measured at angle $\theta \approx 3.3°$ and total light intensity $I \approx 275\,\mathrm{mW/cm^2}$

strength $\gamma\ell$ from 0.9 to 0.95. This gives us for $r = 1$ the ultimate phase-conjugate reflectivity R_{pc} from 1.6 to 1.9.

Finally, we measure the intensity dependence of the oscillation intensity (Fig. 6a) to compare it with the intensity dependence of the phase-conjugate reflectivity (Fig. 6b). As one can see, the threshold intensity of the pump wave is about $100\,\mathrm{mW/cm^2}$. Above the threshold the oscillation intensity increases and saturates at the level of 10% of the total intensity of two pump beams. Note that, at $I = 100\,\mathrm{mW/cm^2}$, the measured phase-conjugate reflectivity corresponds roughly to one-half of its ultimate saturated value. Thus, in spite of the fact that the absolute values of R_{pc} measured with the signal beam are much smaller than estimated from the experiment with coherent oscillation, the ratio of the threshold R_{pc} and saturated R_{pc} is the same in both cases ($0.3/0.17 \approx 1.76$ for BWFWM and $1.7/1.0 \approx 1.7$ for a coherent oscillator).

This comparison suggests the idea that the data on R_{pc} measured with BWFWM are reduced by a certain scaling factor with respect to the real values. There may be several reasons for such reduction of measured efficiency; the most important among them is the Gaussian transverse distribution of beam intensity.

For media with a third-order optical nonlinearity the coupling strength is proportional to the intensity, i.e. the transverse distribution of $\gamma\ell$ is a replica of the Gaussian beam profile:

$$\gamma\ell(x, y) = \gamma\ell_0 \exp\left(-\frac{2x^2 + 2y^2}{\omega^2}\right), \tag{3}$$

where $\gamma\ell_0$ is the coupling strength on the top of the Gaussian intensity distribution and 2ω is the Gaussian beam waist at $1/\mathrm{e}^2$ intensity. To account for the effect of a nonuniform intensity distribution on the measured R_{pc} we calculate the normalized average $\bar{R}_{\mathrm{pc}}$:

$$\bar{R}_{\mathrm{pc}} = \frac{\int\limits_{-\infty}^{\infty}\int\limits_{-\infty}^{\infty} R_{\mathrm{pc}}(x, y) \exp\left(-\frac{2x^2+2y^2}{\omega^2}\right) \mathrm{d}x\,\mathrm{d}y}{\int\limits_{-\infty}^{\infty}\int\limits_{-\infty}^{\infty} \exp\left(-\frac{2x^2+2y^2}{\omega^2}\right) \mathrm{d}x\,\mathrm{d}y}, \tag{4}$$

where $R_{\mathrm{pc}}(x, y)$ is given by (1) and (3). The data for plane waves and for Gaussian beams are shown in Fig. 7a by thick

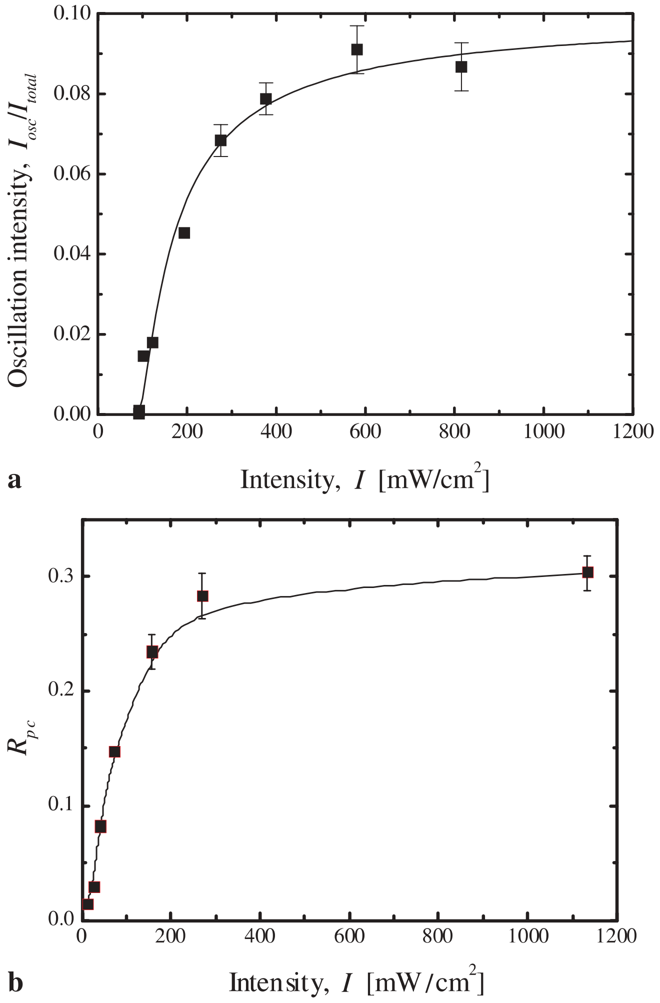

FIGURE 6 Intensity dependence of the oscillation intensity (**a**) and intensity dependence of the phase-conjugate reflectivity (**b**), measured at the same angle $\theta \approx 3°$ with intensity ratio of the pump beams $r \approx 1$

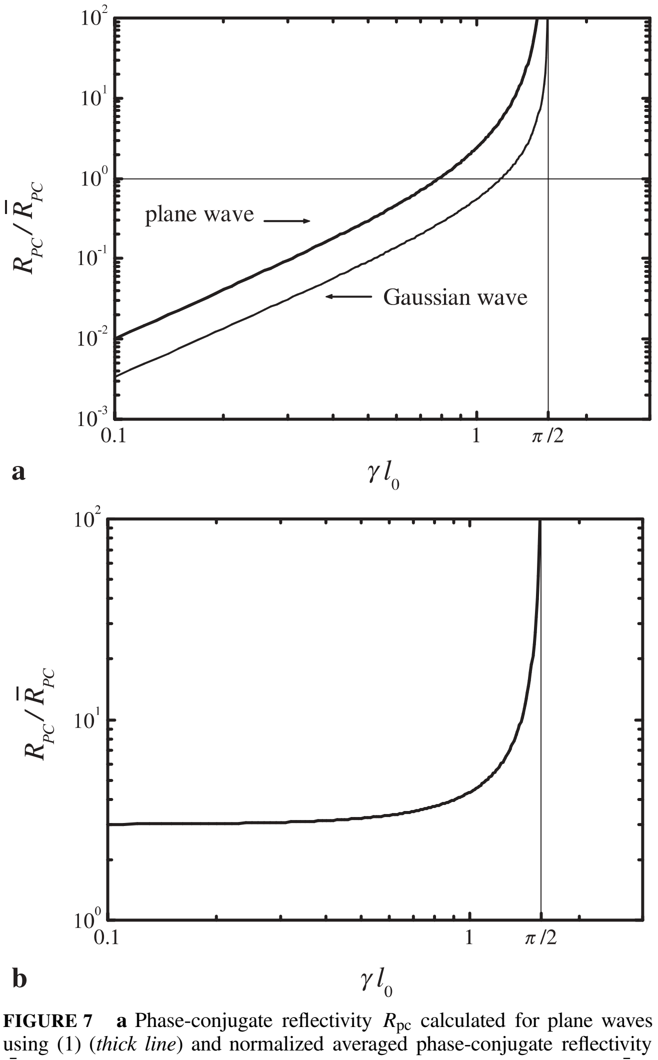

FIGURE 7 **a** Phase-conjugate reflectivity R_{pc} calculated for plane waves using (1) (*thick line*) and normalized averaged phase-conjugate reflectivity $\bar{R}_{\mathrm{pc}}$ calculated for Gaussian beams using (4) (*thin line*). **b** The ratio $R_{\mathrm{pc}}/\bar{R}_{\mathrm{pc}}$, where R_{pc} and $\bar{R}_{\mathrm{pc}}$ correspond to the plane waves and the Gaussian beams, respectively

and thin lines, respectively. In the wide range of small coupling strengths $\gamma\ell_0$ the average phase-conjugate reflectivity for a Gaussian intensity distribution $\bar{R}_{pc}$ is three times smaller than R_{pc} for plane waves given by (1). With the coupling strength increasing the difference is growing and for $\gamma\ell_0 \geq 1$ (the range where oscillation is possible) the ratio $R_{pc}/\bar{R}_{pc}$ becomes larger than 4 (Fig. 7b).

Therefore, the nonuniform transverse intensity distribution explains (at least in part) the difference of the phase-conjugate reflectivity R_{pc} measured in the BWFWM experiment and estimated from the coherent oscillation experiment.

An additional factor which reduces the measured R_{pc} in BWFWM experiments is incomplete overlap of the signal and pump waves, which can never be absolutely perfect. In the experiment with coherent oscillation the system appears to be self-adjustable; no signal wave is introduced from the outside and the oscillation self-develops with such a spatial structure that profits from the largest possible gain and the highest coupling strength. The beam transverse dimensions are also the subject of self-adjustment: the oscillation beam waist appears to be smaller than that of the pump waves in order to exploit the largest gain near the flat top of the pump wave intensity distribution.

It should be noted that for photorefractive crystals with nonlinearity based on photoinduced charge separation and index modulation via the Pockels effect the effect of a Gaussian intensity distribution of the pump waves on measured values of the phase-conjugate reflectivity is much less dramatic. The reason for this difference is quite clear: within a wide range of light intensity the index modulation in photorefractive crystals is independent of the light intensity.

In conclusion, we report highly efficient (with amplified phase-conjugate reflectivity) and fast (in the subsecond range) backward-wave four-wave mixing in doped CdF_2 crystals with DX-center nonlinearity. Our results prove that wide-band-gap crystals with a center of symmetry like CdF_2 can compete successfully as media for degenerate four-wave mixing with the polar photorefractive crystals.

ACKNOWLEDGEMENTS This study has been supported by the International Science and Technology Center (Project 2136). S. Odoulov is grateful to the Japan Society for the Promotion of Science for an Invitation Fellowship. S. Odoulov and K. Shcherbin cordially thank their colleagues from the Nonlinear Optics Laboratory of Waseda University for hospitality. We are grateful to the reviewers for valuable remarks and suggestions.

REFERENCES

1 A.I. Ryskin, A.S. Shcheulin, E.V. Miloglyadov, R.A. Linke, I. Redmond, I.I. Bushinskaja, P.P. Fedorov, B.P. Sobolev: J. Appl. Phys. **83**, 2215 (1998)

2 R.A. Linke, A.S. Shcheulin, A.I. Ryskin, I.I.Bushinskaja, P.P. Federov, B.P. Sobolev: Appl. Phys. B **72**, 677 (2001)

3 Y. Uesu, K. Yasukawa, N. Saito, S. Itoh, S. Odoulov, K. Shcherbin, A.I. Ryskin: J. Opt. Soc. Am. B **20**, 1905 (2003)

4 J.P. Woerdman: Opt. Commun. **2**, 212 (1970)

5 H.J. Eichler, P. Gunter, D. Pohl: *Laser Induced Dynamic Gratings* (Springer, Berlin, Heidelberg 1996)

6 A. Yariv, D. Pepper: Opt. Lett. **1**, 16 (1977)

7 M. Cronin-Golomb, B. Fischer, J.O. White, A. Yariv: IEEE J. Quantum Electron. **QE-20**, 12 (1984)

8 S. Odoulov, M. Soskin, A. Khyzhnyak: *Optical Coherent Oscillators with Degenerate Four-wave Mixing (Dynamic Grating Lasers)* (Harwood, Chur, London 1991) pp. 37–39

9 J. Feinberg, R. Hellwarth: Opt. Lett. **5**, 519 (1980)

10 A. Shumelyuk, A. Hryhorashchuk, S. Odoulov: Trends Opt. Photon. **87**, 320 (2003)

11 P. Mathey, S. Odoulov, D. Rytz: J. Opt. Soc. Am. B **19**, 2967 (2002)

12 D. Pepper, D. Fekete, A. Yariv: Appl. Phys. Lett. **33**, 41 (1978)

13 E. Le Bihan, P. Varkeerk, G. Grynberg: Opt. Commun. **56**, 202 (1985)

14 L. Richard, J. Maurin, J.P. Huignard: Opt. Commun. **57**, 365 (1986)

15 S. Slussarenko, S. Odoulov, M. Soskin: Sov. J. Quantum Electron. **14**, 589 (1984)

16 A.A. Bagan, V.B. Gerasimov, A.V. Golyanov, V.E. Ogluzdin, V.A. Sugrobov, I.L. Rubtsova, A.I. Khyzhnjak: Sov. J. Quantum Electron. **17**, 49 (1990)

17 P.P. Federov, I.I. Bushinskaja, S.P. Ivanov, B.P. Sobolev, A.S. Shcheulin, A.I. Ryskin: Opt. Spectrosc. **92**, 133 (2001)