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The effect of a photovoltaic field on the Bragg condition for volume holograms in $LiNbO_3$

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Abstract. Experimental and theoretical studies of the photovoltaic shift in the position of the diffraction efficiency maximum of holograms recorded in $LiNbO_3$ for the case of optimal electric field multiplexing are described. The experimental data are explained using a model in which the bulk photovoltaic field is excited in a crystal that is electrically connected with a low loading resistance. We suggest that the surface conductivity of the crystal can play an important role in the formation of this effective loading resistance.

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Photorefractive $LiNbO_3$ crystals exhibit a pronounced photovoltaic effect which consists in a unidirectional motion of the charge carriers excited by illumination of the crystal by light [1, 2]. During holographic recording the photovoltaic effect gives rise to motion of the photo-excited carriers from the illuminated areas toward dark ones, thereby causing the nonhomogeneous charge (and electric field) distribution in the crystal and, hence, formation of a phase hologram through the electro-optic effect. If the sample is not short-circuited, the homogeneous (E_{phv}) component of the field (the bulk photovoltaic effect) appears in addition to the nonhomogeneous space-charge field due the photovoltaic effect. The homogeneous field results from compensation of the photovoltaic current by the conductivity current; it is directed along the c axis of the crystal, and its magnitude depends on the sample conductivity and loading resistance if the resistance is connected to the sample surfaces that are perpendicular to the c axis. In the case of the short-circuit condition (or if the surfaces mentioned above are connected to the outside source of voltage) this field is zero. In the opposite case (open circuit) the field can be almost 100 kV/cm [1]. A sufficiently strong homogeneous photovoltaic electric field can change the average refractive index of the crystal and thus affect the Bragg condition at hologram recording. Remember that the Bragg condition for the grating with spacing Λ is given by

$$\Lambda = \frac{\lambda}{2n \sin\theta} \tag{1}$$

where λ is the light wavelength, θ is the Bragg angle (or half of the angle between the recording beams inside the crystal), and n is the refractive index of the material (in our case n is a function of the electric field).

Very often in holographic experiments the homogeneous field can be ignored or reduced to a necessary low level. However, the situation is different in the case of the so-called electric field multiplexing of volume holograms in $LiNbO_3$ [3].

The technique of electric field multiplexing involves recording of several holograms in the presence of an external electric field. Each hologram is recorded under a definite electric field, and it is expected that that hologram reconstruction will occur under the same external field. This technique is based on a high selectivity of volume holograms, which means that hologram reconstruction has to be performed under the same Bragg conditions as recording. Application of the external electric field to an electro-optic crystal similar to $LiNbO_3$ gives rise to changes in the average refractive index of the material that are included in the Bragg condition. Thus different external fields provide different Bragg conditions, and, therefore, different holograms can be retrieved by merely switching the external electric field rather than changing the wavelength or incident angle as in the spectral or angular multiplexing of volume holograms. The highest electric field selectivity is reached for the reflection geometry of holographic recording (similar to spectral selectivity) [4]. In order to use the technique of electric field multiplexing efficiently, one has to find the optimal orientation of the holographic grating vector, optimal orientation of the external field, and a proper orientation of the readout light polarization. In addition, the reflection geometry should be used. Analysis of the optimal conditions [5–7] showed that the electric field must be applied at a definite angle relative to the c axis (approximately 30–45°). However, in this case the crystal is not short-circuited along the c axis, which means that E_{phv} can arise and make a contribution to the refractive index and Bragg condition. In the electric field multiplexing technique the homogeneous field E_{phv} plays rather a detrimental role because it is not stable in time (it decays after the illumination is switched off) and its value depends on many conditions at recording. Although the influence of this field was detected in many experiments [3, 5, 7], no detailed analysis of the effect has been performed up to now to our knowledge. The

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goal of our work was to study experimentally and theoretically the photovoltaic homogeneous electric field arising at holographic recording under the conditions that are optimal for electric field multiplexing.

1 Theoretical background

Here we present a semi-quantitative analysis, which allows us to build a qualitative model of the phenomenon and explain all major experimental results. A development of a more detailed theory is very cumbersome in our case because our experimental situation is in fact two-dimensional and too complicated for a rigorous analysis. Below, we use a very well-known standard approach of a monopolar photorefractive effect [8], so we omit the comments about the conditions for validity of the model.

The electric scheme consists of the photovoltaic crystal that is illuminated by the interference pattern formed by recording beams, and a loading resistance R_l connected in parallel. It can be represented by an equivalent scheme that consists of a source of current J, an internal resistance R_int, and a loading resistance R_l connected in parallel (see Fig. 1). No external voltage is applied. The direction of current J coincides with the c axis of the crystal. The current in the outside circuit (J_out in R_l) under steady-state condition is

$$J_\mathrm{out} = jS \tag{2}$$

where j is the current density and S is the crystal cross-section. If a simple holographic grating is recorded, then in accordance with the definition of the current density in the outside circuit we obtain (see, for example, [2, 8]),

$$j = e\mu n(x)E(x) - eD\frac{\mathrm{d}n(x)}{\mathrm{d}x} + \alpha G I(x)\,. \tag{3}$$

Here, $I(x) = I_\mathrm{o}[1 + m\cos(kx)]$ is the light intensity illuminating the crystal, m is the contrast ratio, I_o is the average light intensity, $k = 2\pi/\Lambda$ is the grating wave vector (k vector and also the x axis are oriented along the c axis), e is the electron charge, μ is the charge mobility, G is the effective Glass constant which characterizes the photovoltaic effect strength, α is the light absorption coefficient, D is the diffusion coefficient, $E(x)$ is the electric field in the crystal, and $n(x)$ is the density of photo-excited mobile carriers.

The first term in (3) describes the current due to the sample conductivity itself, the second term describes the diffusion current, and the third term describes the photovoltaic current.

In the most general case, the function $n(x)$ is given by $n(x) = n_\mathrm{o}[1 + a\cos(kx + \varphi)] + n_\mathrm{d}$, where the parameters a and φ are must be found from the rigorous solution of the Kukhtarev equations [9, 10], n_d is the charge density due to dark conductivity, $n_\mathrm{o} = wI_\mathrm{o}/\mu e = \sigma_\mathrm{o}/\mu e$, $\sigma_\mathrm{o} = wI_\mathrm{o}$ is the photoconductivity, and w is the coefficient describing the generation rate of the photo-excited carriers. In our consideration we use the approach of linear photogeneration and recombination, and therefore from the point of view of requirements to the photogeneration regime there are no limitations on the magnitude of m. However, it is convenient to take $m < 1$ to simplify mathematical calculations or even $m < 0.3 - 0.5$ to provide the condition $a = m$ which allows us to avoid a rather complicated situation ($a > m$) described in [9, 10]. Since we admit that for heavily doped $LiNbO_3$ crystals the photovoltaic field is less than the so-called saturation field E_q and much stronger than the diffusion field (even for high spatial frequencies), we take $\varphi = 0$. We also ignore the dark conductivity (which means that $n_\mathrm{d} = 0$). Then, using the relationship $n(x) = n_\mathrm{o}[1 + m\cos(kx)]$, we can find quantities J, R_int, and hence output voltage U_out. The value J can be found from the short-circuit condition: $R_\mathrm{l} = 0$, $U_\mathrm{out} = 0$, which means that the homogeneous electric field $\langle E(x)\rangle$ equals 0 or

$$\langle E(x)\rangle = \int_0^T \frac{j - \alpha G I(x)}{e\mu n(x)}\,\mathrm{d}x = 0\,. \tag{4a}$$

Equation (4a) does not include the diffusion current because in the steady-state condition diffusion processes make no contribution to the homogeneous current. Assuming, for simplicity, that the length of the crystal $T = q\Lambda$, where q is an integer, we derive the expression for the total current J

$$J = -\alpha G I_0 S\sqrt{1 - m^2}\,. \tag{4b}$$

Here the sign (–) indicates that J and J_out have different propagation directions. Then we can find the output voltage for the open circuit ($R_\mathrm{l} = \infty$ and $J_\mathrm{out} = 0$). In this case we get

$$U_\mathrm{out}^\mathrm{op.c} = -\frac{\alpha G T}{w}\,. \tag{5}$$

This value is independent of the light intensity because we ignore the dark conductivity.

It follows from (4) and (5) that

$$R_\mathrm{int} = \frac{T}{wSI_0}\frac{1}{\sqrt{1 - m^2}}\,. \tag{6}$$

In general, the homogeneous photovoltaic field is given by

$$E_\mathrm{pvh} = \langle E(x)\rangle = \frac{U_\mathrm{out}}{T} = -\frac{\alpha G S I_0}{T}\sqrt{1 - m^2}\frac{R_\mathrm{int}R_\mathrm{l}}{R_\mathrm{int} + R_\mathrm{l}}\,. \tag{7}$$

It follows from (7) that in the case $R_\mathrm{int} \ll R_\mathrm{l}$ the photovoltaic field does not depend on either the light intensity or contrast ratio. This is quite obvious because we neglected dark conductivity and because the spatial modulation of the photovoltaic current is fully compensated by the spatial modulation of the photoconductivity. In the opposite case, when $R_\mathrm{int} \gg R_\mathrm{l}$, the situation is different and

$$E_\mathrm{pvh} = -\frac{\alpha G S I_0}{T}\sqrt{1 - m^2}R_\mathrm{l}\,. \tag{8}$$

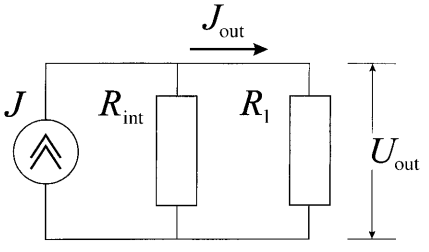

Fig. 1. Equivalent electric scheme of the photovoltaic crystal. J is the source of current, R_int is the internal resistance, R_l is the loading resistance

Here the photovoltaic field depends on both the light intensity and the contrast ratio as well as on the loading resistance. In the following section these dependences will be compared with our experimental data.

2 Electric field shift

The experimental setup is shown in Fig. 2. A cw Nd:YAG laser (1) with intracavity frequency-doubling and an output power of $P \approx 100$ mW at $\lambda = 532$ nm was used. After passing through the beam expander (2) which consists of a microlens, a pinhole and a collimating lens, the light beam was split into two arms, σ_i and ϱ, respectively. The sample (6) was placed between two mirrors (4) and was illuminated from opposite sides by the recording beams σ_i and ϱ. The Bragg angle was 89° 30'. By using a set of gray filters we could vary the power of the recording beams. A single crystal of $LiNbO_3$ doped by Fe^{2+} ions in the concentration less than 0.05 mol % was used, oriented as shown in Fig. 2b. The size T of the sample along the light-propagation direction was 7 mm. A pair of electrodes was deposited on the right and left surfaces, having a distance between the electrodes of 2.5 mm. After hologram recording, beam σ_i was blocked by a shutter (5), and the reconstructed beam σ was detected by a photodiode (7). In most cases, recording was carried out without any applied electric field ($E_0 = 0$). However, during reconstruction, an external field was applied, and the dependence of the diffraction efficiency of the reconstructed hologram on the applied electric field E_0 was measured. Figure 3 shows one of the examples of this dependence. These data were obtained 10 min after hologram recording. The recording conditions were as follows: the external electric field was set to zero, $E_0 = 0$, the duration of recording (t_R) was 10 s, and the intensity of the recording light I_R was 688 mW/cm^2. Figure 3 shows clearly that the maximum diffraction efficiency was achieved under the external field of 0.75 kV/cm, i.e. that the position of the diffraction efficiency maximum was shifted towards higher E_0 compared

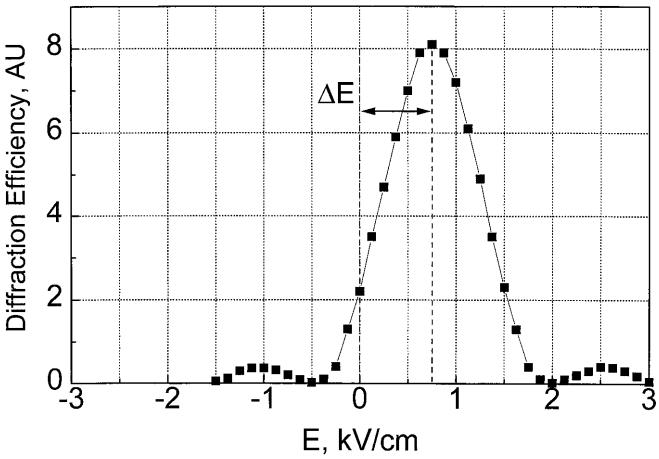

Fig. 3. Diffraction efficiency of a single recorded hologram versus the external electric field. The hologram was recorded at the electric field $E_R = 0$. The electric field shift is $\Delta E = E_{max} - E_R \approx 0.75$ kV/cm

with recording conditions. We define the electric field shift ΔE by the difference between the magnitude of the electric field that has to be applied to the crystal to obtain the maximum diffraction efficiency at reconstruction (E_{max}) and the magnitude of the electric field applied at recording (E_R), i.e. $\Delta E = E_{max} - E_R$. In addition, experiments were performed with the external field applied during recording; we found that if parameters t_R and I_R are fixed, the shift ΔE does not depend on E_0. Moreover, the sign of the shift depends on the orientation of the c axis. For two different crystal orientations shown in Figs. 2a and b where the c axis is oriented in opposite directions, but the external field has a fixed orientation, the shift has opposite signs. The magnitude of the shift does not depend on whether electrodes were short-circuited or not during recording. Figure 4 shows ΔE as a function of the time after hologram recording for different recording beam intensities but a nearly constant exposure energy of 6.88 J/cm^2. During readout the readout beam was switched

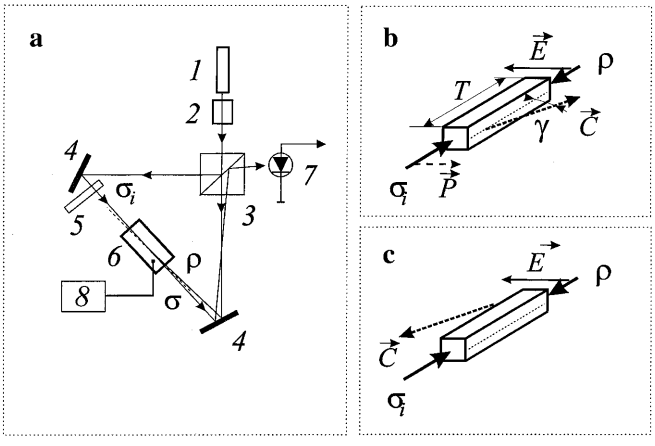

Fig. 2. a Experimental setup. 1, Nd:YAG cw 'ADLAS' laser, $\lambda = 532$ nm; 2, beam expander; 3, beamsplitter; 4, mirrors; 5, shutter; 6, crystal with electrodes; 7, photodiode; 8, tunable high-voltage source. **b** Orientation of the $LiNbO_3$ crystal. ϱ, σ_i are the recording beams, γ is the angle between the optical axis and the direction of the light propagation, $\boldsymbol{P}$ is the orientation of the light polarization, $\boldsymbol{E}$ is the external electric field, T is the thickness of the crystal. **c** The opposite direction orientation of the $\boldsymbol{C}$ axis: the crystal is rotated through 180°

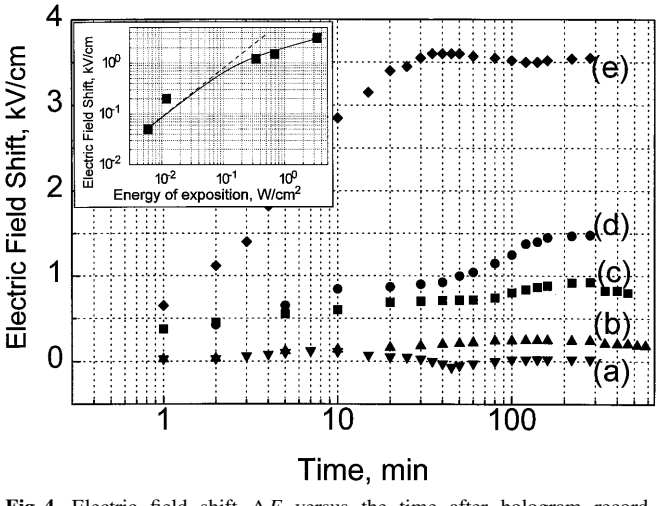

Fig. 4. Electric field shift ΔE versus the time after hologram recording. The contrast ratio is $m \approx 0.95$. (a) $t_R = 1200$ s; $I_R = 0.006$ W/cm^2; (b) $t_R = 600$ s; $I_R = 0.012$ W/cm^2; (c) $t_R = 20$ s; $I_R = 0.344$ W/cm^2; (d) $t_R = 10$ s; $I_R = 0.688$ W/cm^2; (e) $t_R = 2$ s; $I_R = 3.4$ W/cm^2. *Insert:* Electric field shift versus the energy of exposition. The time after hologram recording is 200 min

on for a short period, and the electric field was applied for the same period to measure the diffraction efficiency maximum. The rest of the time the crystal was in the darkness, and no electric field was applied. In the high-power (short-time) recording regime we achieve ΔE of up to 3.5 kV/cm. One can see that after 60–100 min ΔE reaches a 'saturation' level, and the magnitude of ΔE varies approximately linearly with the recording light intensity at low intensities (Fig. 4). Figure 5 shows the dependence of ΔE on the contrast ratio of the recording light m. During measurements of this dependence, the total intensity was kept constant because variations in the total intensity could result in the errors in the dependence of ΔE. The solid curves are theoretical calculations using (8). Note that (8) was obtained for the case $m < 1$. Thus the discrepancy between theory and experiment near $m = 1$ is not surprising. For the case of $m < 0.6$, the experimental data and the theoretical predictions were found to be in rather good agreement.

The obtained experimental results can be explained in the following way. During recording, the photovoltaic electric field arises and modifies the average refractive index and Bragg conditions in the same manner as an external electric field. After recording, the photovoltaic field relaxes, and during reconstruction one has to apply an additional external electric field to compensate for the refractive index changes caused by the photovoltaic field which existed during recording. (In our geometry the directions of the photovoltaic and applied fields differ by an angle of $90° - \gamma = 60°$.) This is the origin of the shift. This model agrees qualitatively quite well with our experimental data. The shift is proportional to the light intensity I_0, it decreases with increasing m, and changes its sign if the direction of the c axis is reversed. This model can explain even why the shift changes the sign if recording is performed at a very low intensity (see Fig. 4, data 'a'). In this case, the photovoltaic field that arises during reconstruction can exceed the weak photovoltaic field at recording, this resulting in the shift of the opposite sign. The relaxation time of the homogeneous photovoltaic field is determined by electric leakage (or effective resistance R_l) and is much shorter than the typical dielectric relaxation time for this crystal ($\tau_M \approx 10^5$–10^6 s) which confirms the assumption that $R_l \ll R_{int}$. Therefore, the model which includes a loading resistance that is independent of the light intensity (for the case where the condition $R_l \ll R_{int}$ is fulfilled) explains all major experimental data although no loading resistance is connected to the crystal in reality. A rigorous analysis of the possible mechanism of conductivity in the crystal that results in the same effect as the loading resistance is rather complicated because of the two-dimensional character of the problem. Our guess is that the conductivity of the surfaces that are not covered by electrodes and thin subsurface layers may play an important role in our effect. First, very qualitative experiments, in which a reduction of the shift was detected if the crystal was placed in very wet atmosphere are in agreement with this assumption. The fact that the speed of holographic recording and the speed of relaxation of the photovoltaic shift are approximately equal at low intensities of the recording light is also in agreement with our assumption about the important role of the surface conductivity. However, it is not valid for high intensities as long as the condition $R_l \ll R_{int}$ is not valid any more. Nevertheless other mechanisms are possible. We can estimate the absolute value of the homogeneous photovoltaic field by using the experimental value of the shift. Since the photovoltaic field is directed along the c axis, but the external field compensating variation in the average refractive index is directed at angle $90° - \gamma$ to the axis, to get a very approximate estimate, the relationship between the photovoltaic field (E_{phv}) and shift can be derived by taking into account the dependence of the efficient electro-optic effect on the electric field orientation [6]

$$E_{phv} = \Delta E \frac{r_{13}\sin\gamma\cos^2\gamma + r_{33}\sin^3\gamma + 2r_{51}\sin\gamma\cos^2\gamma - r_{22}\cos^3\gamma}{r_{13}\cos^2\gamma + r_{33}\sin^2\gamma}. \quad (9)$$

Here, $r_{13} = 8.6$ pm/V, $r_{33} = 30.8$ pm/V, $r_{51} = 28.0$ pm/V, $r_{22} = 4.3$ pm/V are the electro-optical coefficients for $LiNbO_3$ [10]. For the experimental values of $\gamma = 30°$ and $\Delta E = 1$ kV/cm, we set $E_{phv} \approx 1.79$ kV/cm. This value is much less than that published in [1], but is in agreement with the assumption that the crystal is connected in parallel with a low loading resistance. Because of the low photovoltaic field, the role of the second-order factors in the photovoltaic effect, as, for example, the role of the anisotropy of the Glass constant, were not taken into account up to now. However, a preliminary analysis has shown that these effects are not significant.

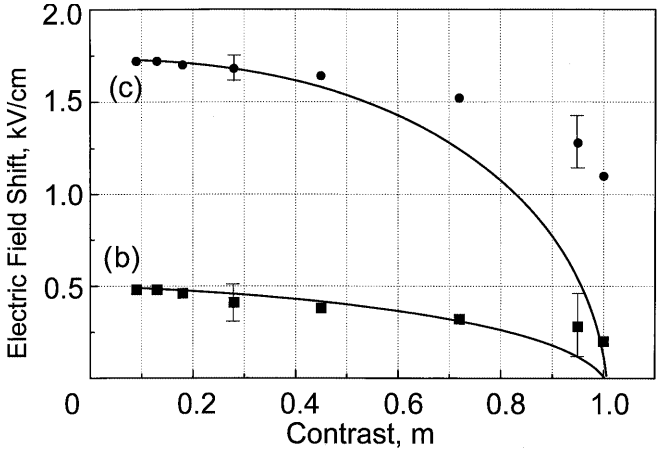

Fig. 5. Electric field shift versus the contrast m. The conditions of recording are (b) and (c) from Fig. 4. *Solid curves* are calculations according to (8)

3 Conclusions

An experimental study of the shift in the position of the diffraction efficiency maximum for holograms recorded in $LiNbO_3$ in an optimal geometry for electric field multiplexing has been carried out. We found that the shift increases linearly with light intensity, reduces with increasing contrast ratio of the interference pattern, and also changes its sign when the c axis reverses its orientation. These data are explained by using a simple theoretical model in which the bulk photovoltaic field is excited in the crystal that is electrically connected with a low loading resistance. Moreover, we suggest that the surface conductivity of the crystal can play

an important role in the formation of the effective loading resistance.

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References

1. A.M. Glass, D. von der Linde, T.J. Negran: Appl. Phys. Lett. **25**, 233 (1974)
2. R. Orlowski, E. Kraetzig: Solid State Commun. **27**, 1351 (1978)
3. M.P. Petrov, S.I. Stepanov, A.A. Kamshilin: Ferroelectrics **21**, 631 (1978); Opt. Commun. **29**, 44 (1979)
4. A. Kewitch, M. Segev, A. Yariv, R.R. Neurgaonkar: Opt. Lett. **18**, 534 (1993)
5. M.P. Petrov, A.V. Shamray, V.M. Petrov, J. Sanchez Mondragon: Opt. Commun. **153**, 305 (1998)
6. A.V. Chamrai, M.P. Petrov, V.M. Petrov: Opt. Soc Am. TOPS Vol. 27, *Advances in Photorefractive Materials and Devices*, p. 515 (Elsinore, Denmark 1999)
7. V.M. Petrov, C. Denz, A.V. Shamray, M.P. Petrov, T. Tschudi: Appl. Phys. B **71**, 43 (2000)
8. N. Kukhtarev, V. Markov, S. Odulov, M. Soskin, V. Vinetskii: Ferroelectrics **22**, 949 (1979)
9. N.V. Kukhtarev, P. Buchhave S.F. Lyuksyutov: Phys. Rev. A **55**, 3133 (1997)
10. N.V. Kukhtarev, S.F. Lyuksyutov, P. Buchhave, T. Kukhtareva, K. Sayano, P. Banerjee: Phys. Rev. A **58**, 4051 (1998)