RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 52, NUMBER 4 OCTOBER 1995

# Brillouin scattering and dynamical diffraction of entangled photon pairs

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(Received 27 June 1995)

Photons incident on a periodic medium at the Bragg angle are dynamically diffracted and exhibit the *pendellösung* phenomenon familiar from x-ray and neutron-diffraction experiments. The entangled two-photon state incident from the positive and negative Bragg angle exhibits a *pendellösung* length that is half that for the single-photon case. In our experiment the photon pairs were produced via parametric down-conversion and the periodic medium was a crystal whose refractive index was periodically altered by an acoustic wave. The predicted *Pendellösung*-length behavior was experimentally confirmed.

PACS number(s): 42.50.−p, 03.65.−w, 42.50.Dv, 42.79.Jq

When visible photons enter a medium with a periodic structure under the Bragg condition, dynamical diffraction occurs in a way similar to what has been observed in previous experiments with neutrons [1,2] and x rays [3] diffracted by large perfect crystals. While in kinematic diffraction the beams diffracted by each small volume element are treated as being incoherent, dynamical diffraction theory takes into account all coherent wave superpositions within the whole crystal. The wave fields are coherently coupled, giving rise to a variety of phenomena, such as, for example, anomalous absorption and "*Pendellösung*" [4].

All dynamical diffraction experiments have thus far simply measured the intensities of the diffracted beams, i.e., they have been "first-order" interference experiments. The question now arises as to what kinds of new phenomena will occur if we study the dynamical diffraction of more complicated states; e.g., those containing more than one particle. One then looks for correlations between two or more detection events.

The only correlated states with high intensities presently available in the laboratory are those of pairs of correlated photons produced either by atomic cascades or in the process of parametric down-conversion. Since in the present paper we study a diffraction phenomenon, the latter process is far superior because it provides the photon pairs with well-defined relative momenta. In our experiment, down-converted photons were directed on a periodic medium. We chose diffraction from the periodic refractive-index variations produced by an acoustic wave inside a crystal (such devices are commonly called acousto-optic modulators): due to the photoelastic effect, the acoustic wave alters the refractive index of the crystal, creating a three-dimensional moving index grating whose exact features depend on the shape of the sound wave. In particular, the grating possesses a period $\Lambda$ equal to the wavelength of the sound and travels with the acoustic velocity $v_{ac}$ through the crystal. The resulting interaction between the electromagnetic radiation and the acoustic wave, i.e., between the photons and the phonons, represents one of the most elementary examples of Brillouin scattering. It is convenient to analyze the diffraction phenomena in a reference frame comoving with the sinusoidal index grating at a velocity equal to the velocity of sound in the medium; our quantities will henceforth be defined in that frame. The principle of our experiment is shown in Fig. 1.

The two-photon state incident on the index grating can be written as

$$|\psi_{in}\rangle = \frac{1}{\sqrt{2}}(|\theta_B\rangle_1|-\theta_B\rangle_2+|-\theta_B\rangle_1|\theta_B\rangle_2), \tag{1}$$

where the ket $|\theta_B\rangle$ ($|-\theta_B\rangle$) describes a photon incident at the positive (negative) Bragg angle. Physically, the state of Eq. (1) describes two incident photons, both symmetrical fulfilling the Bragg condition ($\sin\theta_B=\lambda/2\Lambda$, where $\lambda$ is the photon wavelength), and the state has been symmetrized because it is not known which of the two photons is incident on which side. In principle, one could now write a full Hamiltonian describing the medium and solve the wave equation for the state of the down-converted photon pair represented in Eq. (1). However, a much simpler approach suggests itself when one realizes that the solutions for each of the two in-

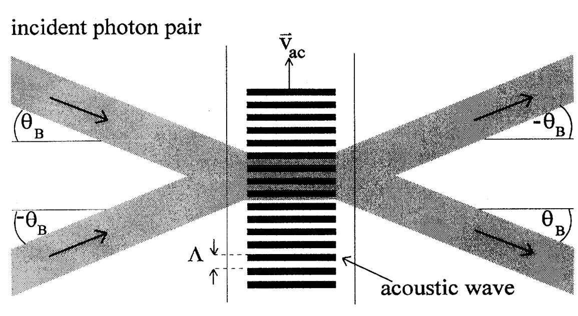


FIG. 1. An acoustic wave propagating through a crystal causes a periodic change in its refractive index, thus creating an index grating that travels with the velocity of sound $v_{ac}$ and has a period $\Lambda$ equal to the acoustic wavelength. The two conjugate photons of a photon pair are directed onto this acousto-optical periodic medium at the Bragg angle and are dynamically diffracted by the induced grating.

 

cidence directions are already well known from dynamical diffraction theory. In fact, when the input state illuminates only one of the two input directions, aside from a common irrelevant phase factor the output state is given by

$$|\theta_B\rangle \rightarrow \cos\left(\frac{\pi l}{l_p}\right)|\theta_B\rangle - i\,\sin\left(\frac{\pi l}{l_p}\right)|-\theta_B\rangle,$$
$$|-\theta_B\rangle \rightarrow \cos\left(\frac{\pi l}{l_p}\right)|-\theta_B\rangle - i\,\sin\left(\frac{\pi l}{l_p}\right)|\theta_B\rangle, \tag{2}$$

where the cosine and sine terms give the probability amplitudes for finding the photons in the corresponding state, and $l$ is the interaction length (in our case the width of the acoustic wave). We see that under the above conditions the intensities of the outgoing beams oscillate with a frequency depending on the characteristic parameter $l_p$, the *Pendellösung* length. Physically, the *Pendellösung* length is that interaction length within the crystal after which a diffracted photon is diffracted back into its initial direction. In a quantum picture it is that crystal length the photon has to transverse in order to return to its initial state. The *Pendellösung* length is a function of various parameters and can immediately be obtained by comparing Eq. (2) with the well-known reflectivity of acousto-optic modulator crystals [5],

$$R \equiv \frac{I_d}{I_{\text{tot}}} = \sin^2\left[\frac{\pi l}{2\lambda_0}\left(\frac{n^6 p^2}{\rho v_{\text{ac}}^3} I_{\text{ac}}\right)^{1/2}\right] = \sin^2(\pi l/l_p), \tag{3}$$

so that

$$l_p = 2\lambda_0 \left(\frac{\rho v_{\text{ac}}^3}{n^6 p^2 I_{\text{ac}}}\right)^{1/2}, \tag{4}$$

and analogously the transmittivity is given by $T=\cos^2(\pi l/l_p)$. $I_d$ is the diffracted intensity and $I_{\text{tot}}$ is the total incident intensity; $v_{\text{ac}}$ is the velocity of sound and $I_{\text{ac}}$ the acoustic intensity inside the crystal; $n$ is the refractive index, $\rho$ the mass density, and $p$ the photoelastic coefficient of the crystal; and $\lambda_0$ is the optical wavelength in vacuum.

To calculate the probability amplitudes (and consequently the intensities of the outgoing beams) for the incident two-photon state of Eq. (1), we simply insert the result of Eq. (2) into Eq. (1) and obtain

$$\begin{aligned} |\psi_{\text{in}}\rangle &= \frac{1}{\sqrt{2}}(|\theta_B\rangle_1|-\theta_B\rangle_2 + |-\theta_B\rangle_1|\theta_B\rangle_2) \rightarrow |\psi_{\text{out}}\rangle \\ &= \frac{1}{\sqrt{2}}[\cos(2\pi l/l_p)(|\theta_B\rangle_1|-\theta_B\rangle_2 + |-\theta_B\rangle_1|\theta_B\rangle_2) \\ &\quad - i\,\sin(2\pi l/l_p)(|\theta_B\rangle_1|\theta_B\rangle_2 + |-\theta_B\rangle_1|-\theta_B\rangle_2)]. \end{aligned} \tag{5}$$

This equation describes the superposition of two possibilities: either both photons leave the index grating in the same output beam (with a probability amplitude given by the sine), or they leave by the two different output beams (the cosine term). Consequently, if we insert a detector into each outgoing beam, the probability for a coincidence count (one photon in each detector) is given by

$$P(1,1) = \cos^2(2\pi l/l_p). \tag{6}$$

The coincidence count rate again varies sinusoidally, analogous to the singles count rates, but *twice* as fast. The *Pendellösung* length for a two-photon state is just half the *Pendellösung* length for individual photons.

The above equations were obtained by assuming monochromatic fields. A more realistic description employing finite photon wavepackets gives the same results as long as the two photons arrive at the crystal simultaneously (i.e., as long as the spatial and temporal overlap of the photons at the crystal is maximal). This can be achieved by adjusting the relative optical path-length difference for the two photons to zero, $\Delta s=0$. Then the amplitudes of the two alternative processes in which there is one photon in each detector (leading to a coincidence count) interfere destructively [6]. On the other hand, for a relative path-length difference greater than the coherence length of the individual photons, one has no interference and the photons behave independently. A measurement of the coincidence count rate as a function of $\Delta s$ therefore reveals a minimum, where the width of the two-photon correlation dip is a measure of the coherence length of the photons. The (normalized) minimum coincidence count rate—corresponding to the simultaneous arrival of the photons ($\Delta s=0$)—is given by Eq. (6). By adjusting the power of the acoustic wave we vary the *Pendellösung* length. Hence in the case of a crystal thickness given by an odd multiple of $l_p/4$, the coincidence count rate at $\Delta s=0$ vanishes.

Note also that due to the moving index grating, the frequency of the photon—when diffracted—is Doppler shifted by an amount equal to the acoustic frequency $\Omega_{\text{ac}}$. This can be understood by considering a light field reflecting off a mirror moving with the acoustic velocity. One might think that this frequency shift would destroy the interference. However, in our experiment the frequency of the acoustic wave was about $10^5$ times smaller than the bandwidth of the photons and thus the influence of the frequency shift can be neglected.

Our experimental setup is shown in Fig. 2. The 2-mm beam of a single-mode argon-ion laser operating at 351.1 nm was directed on a $\chi^{(2)}$ nonlinear crystal ($LiIO_3$), giving rise to pairs of correlated photons simultaneously produced in the process of parametric down-conversion. We selected photons with center wavelengths at 702.2 nm by irises and 5 nm [full width at half maximum (FWHM)] interference filters placed in front of the detectors. At incidence angles of about +8 and −8 mrad, the two conjugate photons of each pair were overlapped at the crystal. They were detected in the two output beams with silicon avalanche photodiodes operating in the Geiger mode. After amplification and pulse shaping, the counts were registered individually as well as fed to a time-to-amplitude converter arrangement to allow coincidence counting. Counts registered within a 15-ns gate window were treated as coincident.

In the experiment we used a crystal (1.8 cm long) of lead molybdate ($PbMoO_4$), driven by a piezoelectric transducer on which an rf signal was applied. By altering the frequency of the signal (and consequently the acoustic wavelength $\Lambda$), we varied the Bragg angle so as to fine tune for the incidence directions of the photons: we counted the photons

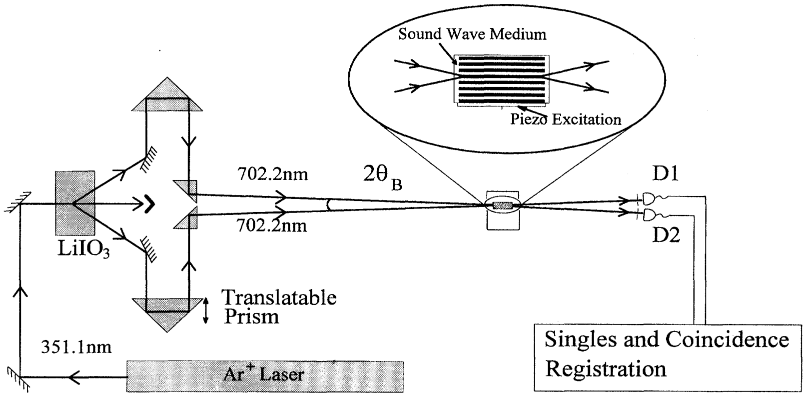


FIG. 2. Experimental setup: down-converted photon pairs are produced in a $LiIO_3$ nonlinear crystal and are directed on an acousto-optical modulator from opposite sides at angles $\pm\theta_B$ (as measured in the rest frame of the traveling sound wave). The relative optical path length of the input arms can be altered by a translatable prism and thus the photons can be made to arrive simultaneously. After passing through irises and interference filters the photons are finally detected by two detectors, one placed in each of the output beams.

in coincidence and maximized the visibility of the resulting dip, which depends sensitively on the overlap of the exit beams and hence on how accurately the two conjugate photons simultaneously fulfill the Bragg condition. The driving frequency thus used was 83.6 MHz corresponding to a Bragg angle of 8.08 mrad and an acoustic wavelength inside the crystal of $\Lambda = 45.4$ $\mu$m.

To examine first the *Pendellösung* phenomenon for single photons we blocked one of the two input arms and detected the transmitted and reflected photons as a function of the acoustic power. In determining the transmittivity and reflectivity it is necessary to account for the different efficiencies of the two detectors. This was done by summing the counts of both detectors for some values of the reflectivity and comparing them. With the irises placed in front of the detectors we selected the photons with an angular divergence of 0.35 mrad, corresponding to a 1-nm bandwidth.

Figure 3(a) shows the normalized singles count rates (for one of the input directions) after subtraction of the background rate, versus the square root of the acoustic power. The rates display the expected sinusoidal oscillations for powers $P < 0.8$ W (higher acoustic powers lead to distortions of the grating), with periodicities of $1.533 \pm 0.027$ $W^{-1/2}$ and $1.566 \pm 0.053$ $W^{-1/2}$ for each input arm, respectively, except for a reduced contrast; this is in reasonable agreement with theory [Eq. (3) predicts a periodicity of 1.54 $W^{-1/2}$]. The maximum reflectivities for each input direction were 80% and 88%, respectively. The discrepancy from 100% can be attributed to a slight misalignment and an imperfect grating (the first maximum in the reflectivity is reached for an acoustic power of $P = 0.95$ W, which is above the onset of distortion). The bandwidth of the photons causes at most a reduction of 0.1%.

For the two-photon experiment we employed a translatable trombone prism to alter the path-length difference of the two input arms. As we counted the photons with the two detectors at the outputs in coincidence, the count rate displayed a dip whose visibility we optimized by slightly translating the crystal to achieve a maximal transverse spatial overlap of the photons.

We recorded the dips for 14 different values of the acoustic intensity. The widths of the dips were about 220 $\mu$m (FWHM), implying a coherence length of the photons of 190 $\mu$m. The maximum dip visibility (for $l_p = 4l$) was 69.7%. As before, the discrepancy from the theoretical value of 100% can be attributed to an imperfect alignment and to inhomogenities of the acoustic wave. We also examined theoretically the dependence of the visibility on slightly different center frequencies for the photons of a pair, as might arise due to

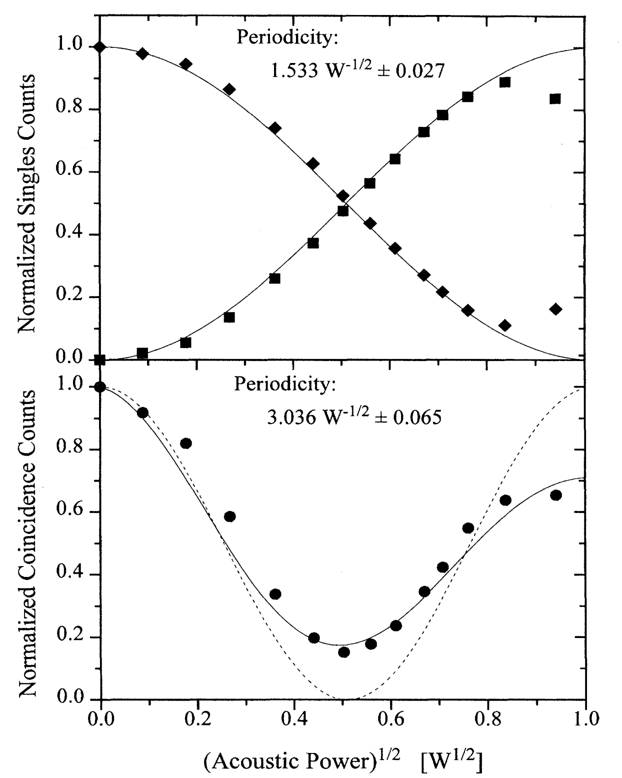


FIG. 3. (a) Photons incident from one input beam only: singles count rates for the two outgoing beams plotted versus the square root of the acoustic power. The curve is a theoretical plot based on Eq. (3). (b) Photons incident in pairs, one in each input beam: coincidence count rate between the detectors placed in the two output beams versus the square root of the acoustic power. The dashed curve is the theoretical prediction according to Eq. (6) and the solid one includes an exponential damping factor (fit to the data) for the cosine modulation in order to accommodate experimental imperfections. The oscillations vary with a period of 3.036 $W^{-1/2} \pm 0.065$, which is twice the periodicity observed in the singles count rates (1.533 $W^{-1/2} \pm 0.027$).

slightly tilted interference filters. We found that frequency deviations of $\pm 0.35$ nm (which is within the accuracy of the filters) would cause already a 30% reduction in visibility.

Figure 3(b) shows the normalized coincidence count rate (for $\Delta s = 0$) versus the square root of the acoustic power. The counts vary according to the cosine-square law (for acoustic powers $P < 0.8$ W) with a periodicity of $3.036 \pm 0.065\ \mathrm{W}^{-1/2}$. This is twice the periodicity obtained for the singles counts, in agreement with the theoretical prediction of Eq. (6).

In conclusion we would like to point out that the present work opens up two new fields of experimentation with entangled photons. These are dynamical diffraction studies on the one hand and Brillouin scattering on the other. In both we expect a rich field for future experimentation.

This work was supported by the Austrian "Fonds zur Förderung der wissenschaftlichen Forschung" (Project No. S6502 and Lise Meitner Fund, Grant No. M077-PHY) and the National Science Foundation (Grant No. PHY92-13964).

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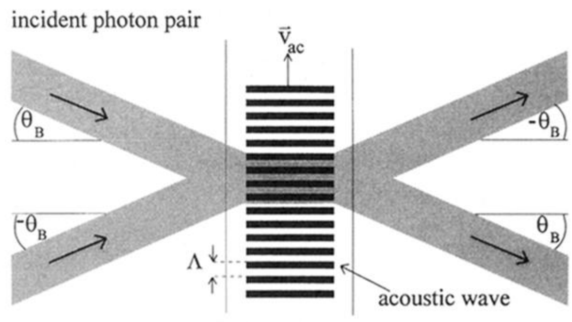


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