Propagation of femtosecond light pulses in a dye solution : Nonadherence to the conventional group velocity

Kohmoto, Toshiro
Fukui, Y.
Furue, S.
Nakayama, K.
Fukuda, Yukio

(Citation)
Physical Review E,74(5):56603-56603

(Issue Date)
2006-11

(Resource Type)
journal article

(Version)
Version of Record

(URL)
https://hdl.handle.net/20.500.14094/90000234

PHYSICAL REVIEW E **74**, 056603 (2006)

Propagation of femtosecond light pulses in a dye solution: Nonadherence to the conventional group velocity

T. Kohmoto,[1] Y. Fukui,[2] S. Furue,[2] K. Nakayama,[2] and Y. Fukuda[1]
[1]*Department of Physics, Faculty of Science, Kobe University, Kobe 657-8501, Japan*
[2]*Graduate School of Science and Technology, Kobe University, Kobe 657-8501, Japan*

(Received 19 August 2006; published 3 November 2006)

Propagation of femtosecond light pulses in a resonant absorption medium is studied. The propagation time of the light pulses was measured in a dye solution by the optical-Kerr-gate method. Nonadherence to the conventional group velocity $d\omega/dk$, which is defined in weak absorption region, was found. The observed wavelength dependences of the group delay and the spectral change in the anomalous dispersion region are qualitatively in good agreement with the theoretical prediction obtained from the new definitions of group velocity, which can be clearly defined even in strong absorption region.

DOI: 10.1103/PhysRevE.74.056603 PACS number(s): 42.25.Bs, 78.47.+p

I. INTRODUCTION

Recently various phenomena on the propagation of light pulses have attracted a lot of interest. Ultraslow propagation [1,2], storage of light [3–6], and superluminal propagation [7–10], have been reported. In these phenomena an artificial processing, the electromagnetically induced transparency [11,12], plays an important role.

However, even in natural resonantly absorbing media, the light propagation is not necessarily understood completely. Conventional group velocity, $d\omega/dk$, is defined in a weak absorption region, in resonantly absorbing media, that can exceed the light velocity in vacuum and even become negative. The study of light pulse propagation through dispersive media started with a series of papers by Sommerfeld and Brillouin [13,14]. They showed that the main signal propagates through the medium with a velocity always less than the light velocity c in vacuum, while the front edge of the precursor can travel at c. Since then the problems on the pulse propagation through resonantly absorbing media have been leaving the physicists to puzzle for a century over the meaning of group velocity in connection with the causality and the theory of relativity.

Analytical and numerical studies of the propagation of a Gaussian-shaped light pulse through a resonantly absorbing medium were reported by Garret and McCumber [15] and Crisp [16]. They gave a remarkable consequence that, when the incident frequency lies in the anomalous dispersion region, superluminal or even negative velocity is possible, but it never violates the causality.

Experimental observation of the negative velocity was reported by Chu and Wong [17] for a thin sample of semiconductor. In their experiment a second-order autocorrelation technique was used to detect the pulses. However, such a technique, as is pointed out by Katz and Alfano [18], cannot reveal any change in pulse shape. A numerical simulation [19] of their data shows that the output pulse is, in fact, strongly distorted, where the concept of pulse velocity has not a clear physical significance.

Recently new definitions [20–22] of group velocity have been proposed, where the distortion of spectral distribution due to the absorption during the pulse propagation is taken into consideration.

Observation of the pulse propagation in a dye solution and comparison with the theoretical results obtained from the new definitions of group velocity were reported by Talukder *et al.* [23]. In their experiment, however, the second-order autocorrelation technique was used again, and wavelength dependence and spectral analysis of the pulse propagation were not reported. To clarify the light propagation in the anomalous dispersion region, more direct and detailed investigation is desired. Recently we observed the negative velocity [24] and the nonadherence to the conventional group velocity [25] for nanosecond light pulses by a direct detection of the pulse intensity in the time domain in natural atomic transitions.

In the present work we studied the propagation of femtosecond light pulses in a dye solution. Methanol solutions of zinc phthalocyanine (ZnPc) are used as the resonantly absorbing media. Wavelength and concentration dependences of the group delay of the optical pulses are measured by the optical-Kerr-gate method. Nonadherence to the conventional definition $d\omega/dk$ of group velocity was observed. The observed group delay and the spectral changes of the transmitted pulses are compared with the theoretical prediction obtained from the new definitions of the group velocity.

II. THEORIES OF GROUP VELOCITY

The conventional group velocity in text books is given by

$$\frac{d\omega}{dk} = \frac{c}{\dfrac{d(n\omega)}{d\omega}} = \frac{c}{n + \omega\dfrac{dn}{d\omega}}. \tag{1}$$

Steep inclination of refractive index, n, in resonantly absorbing regions, can give a large change to the value of $d\omega/dk$ through the contribution of the term $\omega(dn/d\omega)$, even if the value of n is not so changed. The expression of Eq. (1) can be derived under the approximation that the change of $n(\omega)$ is linear around the center frequency ω_c of the light pulse,

$$n(\omega)\omega \simeq n(\omega_c)\omega_c + \left(\frac{d(n\omega)}{d\omega}\right)_{\omega_c}(\omega-\omega_c). \tag{2}$$

The meaning of $d\omega/dk$ is broken down when this approximation is not valid.

Recently new definitions of group velocity, which can be clearly defined even in strong absorption region, have been proposed. The first one is the theory of Tanaka *et al.* [20,21], which was proposed by using the saddle point method. The new group velocity is defined as the average velocity of the pulse peak, taking not only the refractive index but also the spectral change due to the absorption in consideration. The average velocity, which is defined by the traveling distance z of the pulse peak divided by its traveling time t, is given by

$$\frac{z}{t} = \mathrm{Re}\left(\frac{d\omega}{dk}\right)_{\omega_f} = \frac{c}{\left(\frac{d(\mathrm{Re}[n(\omega)]\omega)}{d\omega}\right)_{\omega_f}}, \tag{3}$$

where ω_f is the center frequency of the transmitted pulse. This expression is similar to that of the conventional group velocity. The difference from the conventional group velocity is that the derivative is not taken at the center frequency ω_c of the initial pulse but at ω_f of the transmitted one. If we assume a Gaussian pulse shape, the center frequency of the transmitted pulse is calculated analytically and is given by

$$\omega_f - \omega_c = -\frac{z\delta^2}{c}\left(\frac{d(\mathrm{Im}[n(\omega)]\omega)}{d\omega}\right)_{\omega_f}, \tag{4}$$

where ω_c is the center frequency of the initial pulse, and δ is the spectral width of the initial pulse. The result of this theory says that the group velocity depends on the spectral width or the pulse width of the initial pulse and is determined by the center frequency of the transmitted pulse, which may be shifted from the initial one by the resonant absorption, and that the concept of the conventional group velocity does not break down if applied to the surviving spectrum instead of the initial spectrum.

Another one is the theory of Peatross *et al.* [22]. In their theory, arrival time of pulse is defined by that of the center-of-mass of pulse. The arrival time $\langle t\rangle$ of pulse propagating along the z axis can be expressed as follows:

$$\langle t\rangle = \frac{\int_{-\infty}^{\infty} tS(z,t)dt}{\int_{-\infty}^{\infty} S(z,t)dt}, \tag{5}$$

where $S(z,t)$ is the Poynting vector of the transmitted pulse. Fourier transformation and some calculations lead to a sum of two terms, net group delay and reshaping delay. The reshaping delay has some contribution for frequency-chirped pulses and can be neglected for small frequency chirping. Here we take only the net group delay into account. The net group delay is given by

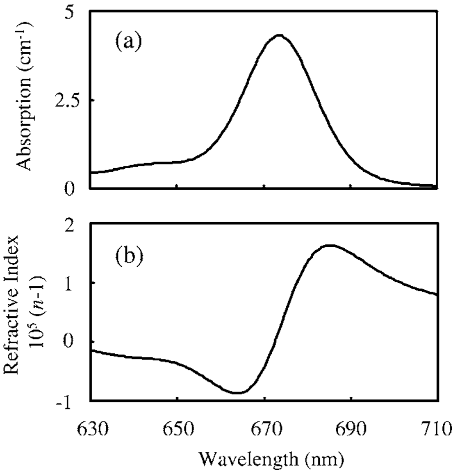

FIG. 1. (a) Absorption coefficient and (b) refractive index change $n-1$ of a methanol solution of zinc phthalocyanine (ZnPc), where the dye concentration is 8×10^{-6} mol/l. The refractive index was calculated from the absorption coefficient observed in the range from 200 to 1100 nm by using the Kramers-Kronig relations.

$$\Delta t = \frac{\int_{-\infty}^{\infty}\frac{d(\mathrm{Re}[k(\omega)])}{d\omega}\Delta z S(z,\omega)d\omega}{\int_{-\infty}^{\infty}S(z,\omega)d\omega} \tag{6}$$

$$= \frac{\int_{-\infty}^{\infty}\frac{\Delta z}{v_g(\omega)}S(z,\omega)d\omega}{\int_{-\infty}^{\infty}S(z,\omega)d\omega}, \tag{7}$$

where $S(z,\omega)$ is the Fourier transform of $S(z,t)$. The arrival time is derived to be the weighted sum of the propagation time for each frequency component of the transmitted pulse. The result of this theory gives a new context for the group velocity, where the group velocity is always meaningful even for broadband pulses and when the group velocity is superluminal or negative, if we consider the function $v_g(\omega) = [d\omega/dk]_\omega$ as group velocity that is permitted to vary arbitrarily within the bandwidth of the pulse.

III. EXPERIMENT

A methanol solution of zinc phthalocyanine (ZnPc) is used as the resonantly-absorbing medium. The absorption and refractive index of the solution are shown in Fig. 1, where the dye concentration is 8×10^{-6} mol/l.

The refractive index was calculated from the absorption coefficient observed in the range from 200 to 1100 nm by using the Kramers-Kronig relations [26]. ZnPc in the solution shows an absorption peak around ~673 nm, and has no other strong absorption peak in the visible and near-infrared regions. The region around the absorption peak is the anomalous-dispersion region, where the frequency derivative of the refractive index becomes negative.

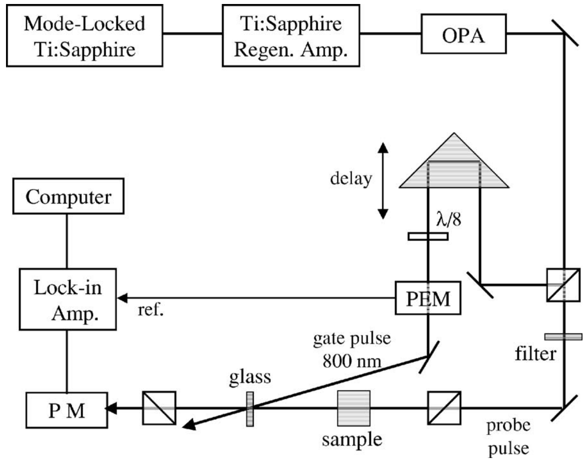

FIG. 2. Experimental setup for the measurement of the propagation time.

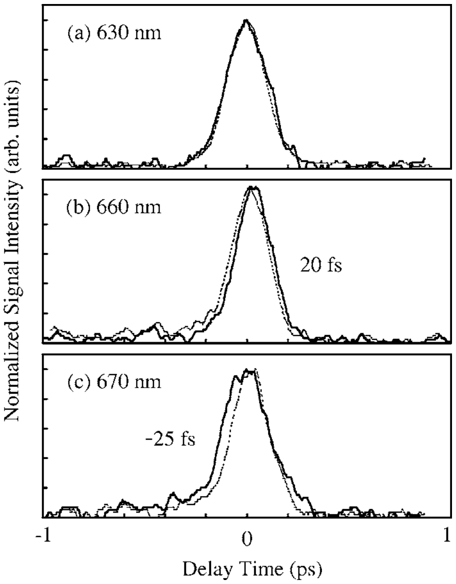

FIG. 3. Wavelength dependence of the observed pulse shape for ZnPc in methanol. The dye concentration is 8×10^{-6} mol/l. The solid curves show the observed transmitted pulse shapes for the dye cell, and the broken curves show those for the reference cell.

Propagation of femtosecond pulses in the dye solution is studied. Figure 2 shows the experimental setup for the measurement of the propagation time. The gate and probe pulses are provided by a Ti:sapphire regenerative amplifier and an optical parametric amplifier (OPA), respectively. Their pulse width is ~150 fs. The probe pulse passes through the dye solution in a glass cell, whose pass length for the probe pulse is 10 mm. Temporal pulse shape after passing through the dye cell is observed by the optical-Kerr-gate method, where an SFS1 glass plate with the thickness of 0.5 mm between crossed polarizers is used as the Kerr medium. SFS1 glass is known to have a large nonlinear refractive index [27]. The linearly polarized gate and probe pulses are nearly collinear and focused on the glass plate. The polarization plane of the probe pulse is tilted by 45° from that of the gate pulse. The time separation between the gate and probe pulses is swept by using an optical delay line. The temporal pulse shape is also observed for a reference cell without dye, which contains only the solvent, and this shape is used as zero delay shape. The delay of the arrival time of the probe pulse after passing through the dye cell from that through the reference cell is measured. This time delay, which we refer to as group delay, is obtained for each wavelength around the absorption peak of ZnPc in methanol. To improve the signal-to-noise ratio, the polarization of the gate pulse is switched between linear and circular polarizations shot by shot by a photoelastic modulator (PEM) with a $\lambda/8$ plate, and the Kerr signal on the probe pulse detected by a photomultiplier (PM) is lock-in detected. A fiber optic spectrometer is used for the observation of the optical spectra of the initial and transmitted pulses.

IV. RESULTS AND DISCUSSION

Wavelength dependence of the observed pulse shape for ZnPc in methanol is shown in Fig. 3, where the dye concentration is 8×10^{-6} mol/l. The solid curves show the observed transmitted pulse shapes for the dye cell, and the broken curves show those for the reference cell. For 630 nm of the initial center wavelength, which is far from the absorption peak, no group delay is found. For 660 nm the probe pulse delays by 20 fs, and for 670 nm the probe pulse advances by 25 fs.

Wavelength dependence of the group delay for three values of the dye concentration is shown in Fig. 4. The solid circles show the observed initial-center-wavelength dependence of the group delay around the absorption peak of the solution. Traveling distance is 10 mm, which is the thickness of the glass cell. The broken curves are the theoretical calculation obtained from the conventional group velocity $d\omega/dk$. The solid curves are the theoretical calculation obtained from the theory of Tanaka *et al.* in Eq. (3). The concentration for Figs. 4(b) and 4(c) is two- and four- times of that for Fig. 4(a). The vertical axis of the group delay is normalized by the conventional group delay. Negative group delay means the light velocity in the dye solution is larger than that in the solvent.

The experimental data deviate from the theories in the longer wavelength region. This may be caused by the fact that the frequency chirping of the probe pulse is large in that wavelength region, while, in the region around the absorption peak, the frequency chirping is small. Here we pay attention to the region around the absorption peak. For the dense solution, as is seen in Fig. 4 the experimental result deviates from the conventional definition $d\omega/dk$, and is explained better by the theory of Tanaka *et al.*

Change of the observed spectrum after the transmission is shown in Fig. 5, where the dye concentration is 8×10^{-6} mol/l. The broken and solid curves show the normalized spectra for the initial and transmitted pulses, respectively. For 630 nm of the initial center wavelength, which is far from the absorption peak, no difference is found between the spectra of the initial and transmitted pulses. For 665 nm of the initial center wavelength, which is at the shorter side of the absorption peak, the center wavelength of the trans-

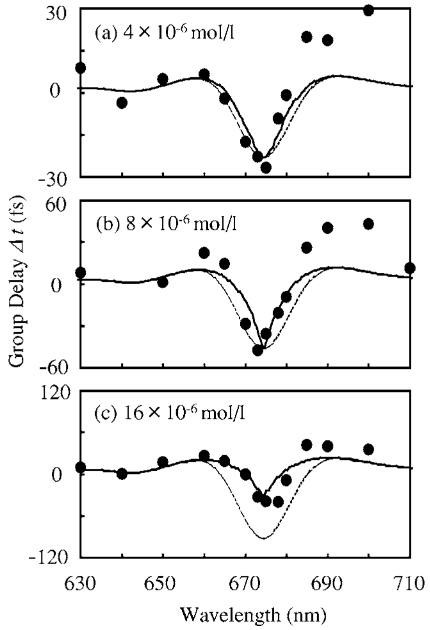

FIG. 4. Wavelength dependence of the group delay for the dye concentration of (a) 4×10^{-6} mol/l, (b) 8×10^{-6} mol/l, and (c) 16×10^{-6} mol/l. The solid circles show the observed initial-center-wavelength dependence of the group delay. The broken curves are the theoretical calculation obtained from the conventional group velocity $d\omega/dk$. The solid curves are the theoretical calculation obtained from the theory of Tanaka *et al.* in Eq. (3). The vertical axis of the group delay is normalized by the conventional group delay.

mitted pulse shifts to a shorter value from that of the initial pulse. For 685 nm, which is at the longer side of the absorption peak, the center wavelength of the transmitted pulse shifts to a longer value.

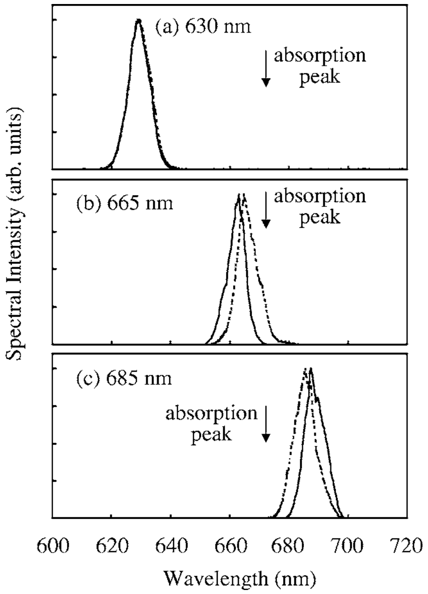

FIG. 5. Change of the observed spectrum after the transmission for the initial center wavelength of (a) 630 nm, (b) 665 nm, and (c) 685 nm. The dye concentration is 8×10^{-6} mol/l. The broken and solid curves show the normalized spectra for the initial and transmitted pulses, respectively.

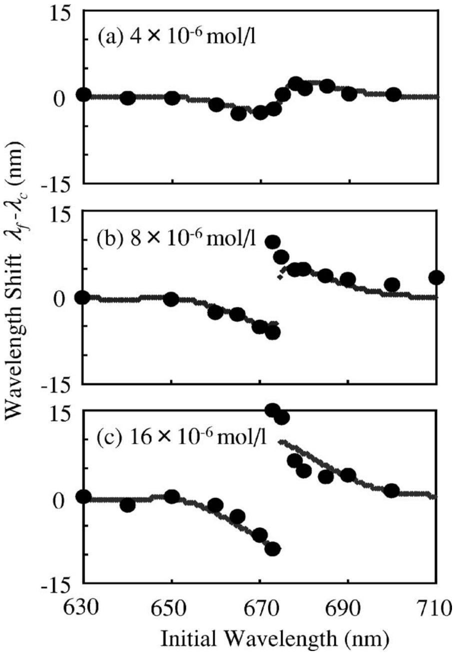

FIG. 6. Wavelength dependence of the wavelength shift for the dye concentration of (a) 4×10^{-6} mol/l, (b) 8×10^{-6} mol/l, and (c) 16×10^{-6} mol/l. The horizontal axis is the center wavelength of the initial pulse and the vertical axis is the shift of the center wavelength of the transmitted pulse from that of the initial pulse. The solid circles show the observed wavelength dependence of the wavelength shift. The solid curves are the theoretical calculation obtained from the theory of Tanaka *et al.* in Eq. (4).

Wavelength dependence of the wavelength shift for the three values of the dye concentration is shown in Fig. 6, where the horizontal axis is the center wavelength of the initial pulse and the vertical axis is the shift of the center wavelength of the transmitted pulse from that of the initial pulse. The solid circles show the observed wavelength dependence of the wavelength shift. Figure 6 shows that the sign of wavelength shift is reversed at the absorption peak, and that the magnitude of the shift becomes larger as the concentration is increased. The solid curves are the theoretical calculation obtained from the theory of Tanaka *et al.* in Eq. (4). As is seen, the experimental result is explained well by the theory of Tanaka *et al.* This wavelength or frequency shift due to the absorption is essential to the deviation from the conventional group velocity.

In the theory of Tanaka *et al.*, the propagation velocity and the frequency shift are derived analytically in Eqs. (3) and (4), and those results are instructive and are easy understand intuitively. However, the calculated frequency dependence of the group delay for asymmetric absorption shapes shows a discontinuous point in the value of group delay at the absorption peak, while it does not appear in the experiment [25].

We also calculated the group delay using the theory of Peatross *et al.*, which does not show discontinuous point even for asymmetric absorption shapes [25]. The solid curves in Fig. 7 show the result of numerical calculation obtained from the theory in Eq. (6).

The theory of Peatross *et al.* also explains the experimental result better than the conventional one. The theoretical

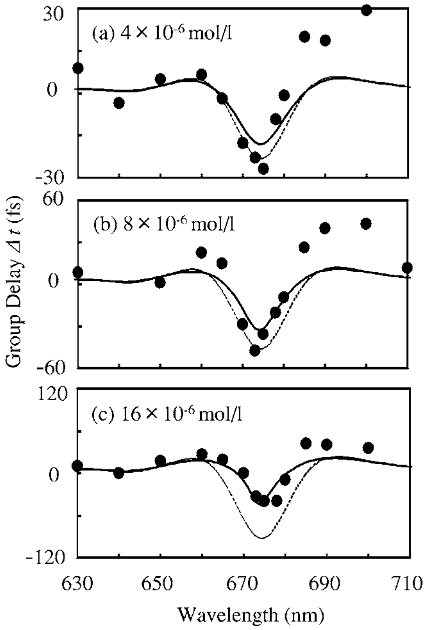

FIG. 7. Wavelength dependence of the group delay for the dye concentration of (a) 4×10^{-6} mol/l, (b) 8×10^{-6} mol/l, and (c) 16×10^{-6} mol/l. The solid circles show the observed initial-center-wavelength dependence of the group delay. The broken curves are the theoretical calculation obtained from the conventional group velocity $d\omega/dk$. The solid curves are the theoretical calculation obtained from the theory of Peatross *et al.* in Eq. (6). The vertical axis of the group delay is normalized by the conventional group delay.

curves in Fig. 7 obtained from the theory of Peatross *et al.* have no discontinuous point not only in the value of group velocity but also in its frequency derivative, while those in Fig. 4 obtained from the theory of Tanaka *et al.* have a discontinuous point in the frequency derivative at the absorption peak. The two theories explain our experimental result better than the conventional one within our experimental error. We cannot say which of the two theories is better to explain our experimental result.

V. SUMMARY

We observed the propagation of optical pulses in dye solutions by using femtosecond laser pulses and the optical-Kerr-gate method, and the experimental results are compared with the new definitions of the group velocity. The group delay was measured in methanol solutions of ZnPc. It was found that the conventional group velocity $d\omega/dk$ is not valid in the strong absorption region. The observed wavelength and concentration dependences of the group delay and the spectral change after the transmission are qualitatively in good agreement with the theoretical prediction obtained from the new definitions of group velocity.

ACKNOWLEDGMENTS

We would like to thank Professor Y. Kanematsu for providing us the SFS1 glass plate.

[1] L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature (London) **397**, 594 (1999).
[2] D. Budker, D. F. Kimball, S. M. Rochester, and V. V. Yashchuk, Phys. Rev. Lett. **83**, 1767 (1999).
[3] C. Liu, Z. Dutton, C. H. Behroozl, and L. V. Hau, Nature (London) **409**, 490 (2001).
[4] D. F. Phillips, A. Fleischhauer, A. Mair, and R. L. Walsworth, M. D. Lukin, Phys. Rev. Lett. **86**, 783 (2001).
[5] A. S. Zibrov, A. B. Matsko, O. Kocharovskaya, Y. V. Rostovtsev, G. R. Welch, and M. O. Scully, Phys. Rev. Lett. **88**, 103601 (2002).
[6] M. Bajcsy, A. S. Zibrov, and M. D. Lukin, Nature (London) **426**, 638 (2003).
[7] L. J. Wang, A. Kuzmich, and A. Dogariu, Nature (London) **406**, 277 (2000).
[8] A. Dogariu, A. Kuzmich, and L. J. Wang, Phys. Rev. A **63**, 053806 (2001).
[9] E. E. Mikhailov, V. A. Sautenkov, I. Novikova, and G. R. Welch, Phys. Rev. A **69**, 063808 (2004).
[10] H. Kang, G. Hernandez, and Y. Zhu, Phys. Rev. A **70**, 011801(R) (2004).
[11] K.-J. Boller, A. Imamoglu, and S. E. Harris, Phys. Rev. Lett. **66**, 2593 (1991).
[12] S. E. Harris, Phys. Today **50**(7), 36 (1997).
[13] A. Sommerfeld, Z. Phys. **8**, 841 (1907).
[14] L. Brillouin, in *Wave Propagation and Group Velocity* (Academic, New York, 1960).
[15] C. G. B. Garrett and D. E. McCumber, Phys. Rev. A **1**, 305 (1970).
[16] M. D. Crisp, Phys. Rev. A **4**, 2104 (1971).
[17] S. Chu and S. Wong, Phys. Rev. Lett. **48**, 738 (1982).
[18] A. Katz and R. R. Alfano, Phys. Rev. Lett. **49**, 1292 (1982).
[19] B. Segard and B. Macke, Phys. Lett. **109A**, 213 (1985).
[20] M. Tanaka, M. Fujiwara, and H. Ikegami, Phys. Rev. A **34**, 4851 (1986).
[21] Y. Amagishi, H. Nakagawa, and M. Tanaka, Phys. Rev. E **50**, 2217 (1994).
[22] J. Peatross, S. A. Glasgow, and M. Ware, Phys. Rev. Lett. **84**, 2370 (2000).
[23] Md. Aminul Islam Talukder, Y. Amagishi, and M. Tomita, Phys. Rev. Lett. **86**, 3546 (2001).
[24] H. Tanaka, H. Niwa, K. Hayami, S. Furue, K. Nakayama, T. Kohmoto, M. Kunitomo, Y. Fukuda, Phys. Rev. A **68**, 053801 (2003).
[25] T. Kohmoto, H. Tanaka, S. Furue, K. Nakayama, M. Kunitomo, and Y. Fukuda, Phys. Rev. A **72**, 025802 (2005).
[26] C. Kittel, in *Introduction to Solid State Physics*, Seventh Edition (Wiley, New York, 1996), p. 308.
[27] R. Nakamura and Y. Kanematsu, Rev. Sci. Instrum. **75**, 636 (2004).