Dissipation-induced effects in the generation of harmonics of a strong elliptically polarized light field in gases

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The general structure of the polarization vector of an isotropic (gaseous) medium at the frequency $n\omega$ in the field of strong elliptically polarized laser light with a frequency ω is analyzed. It is found that in harmonic generation from a strong light field the experimentally discovered rotation of the polarization ellipse of the harmonics with respect to the polarization ellipse of the pump field is caused by the skew-Hermitian (dissipation) part of the generalized nonlinear susceptibilities. The threshold anomalies emerging in the process of harmonic generation are discussed. The polarization dependence of the harmonic yield in an elliptically polarized pump field is analyzed. Finally, the dissipation-induced effect of "elliptic dichroism" is discussed, and it is found that this effect leads, among other things, to a dependence of the yield of the linearly polarized component of a harmonic on the sign of the degree of circular polarization of the pump radiation.
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1. INTRODUCTION

High-harmonic generation of intense optical radiation is one of the most interesting nonlinear optical processes in atomic gases, with important applications, in particular as a promising method for generating short-wave radiation. For instance, in ultrastrong fields with intensities $I>10^{15}$ W cm^{-2} and pico- and femtosecond duration in noble gases, harmonics of neodymium laser light with n up to 141 and a wavelength shorter than 7.5 nm were observed.[1] While the dependence of the yield $I_{n\omega}$ of the nth harmonic on the frequency ω and the intensity I of the pump radiation has been studied in dozens of papers, the polarization effects in the process of harmonic generation remain practically uninvestigated, with linearly polarized pump radiation commonly used in experiments. The first experiments with elliptically polarized pump radiation have been done only recently;[2–5] the polarization of the harmonics was not analyzed and only the dependence of $I_{n\omega}$ on the ellipticity parameter γ (the ratio of the semiaxes of the polarization ellipse) of the pump radiation was studied. Note that the results of the experiments of Liang *et al.*[3] in measuring the dependence of the yield of lowest-order harmonics on γ for $I\leqslant 10^{13}$ W cm^{-2} qualitatively agree with the results obtained by perturbation-theory techniques in Ref. 6.

Recently Weihe *et al.*[7] carried out the first measurements of the polarization characteristics of harmonics in argon for different ellipticity parameters of the pump radiation from a Ti:sapphire laser with an intensity $\sim 10^{15}$ W cm^{-2} and a wavelength 785 nm. The accuracy of the experiment proved insufficient for absolute measurements of the degree of ellipticity of the harmonics but a basic fact was established: the rotation of the principal axis of the polarization ellipse of the harmonics through a certain angle in relation to the principal axis of the polarization ellipse of the pump radiation. This angle is different for different harmonics and increases monotonically with γ. The effect is independent of the density of the gas, which excludes effects of the medium, such as the self-induced rotation of the polarization ellipse, and made it possible for Weihe *et al.*[7] to conclude categorically that the effect is related to the properties of the nonlinear response of a separate atom.

At present there exists no consistent theoretical analysis of the process of harmonic generation in a strong elliptically polarized pump field. The result of this paper indicate that, in contrast to the case of linear polarization, for multiphoton processes in elliptically polarized laser fields it is very important to allow for dissipation effects, which lead to a number of specific polarization phenomena similar to the dissipation-induced circular dichroism in ordinary scattering of light in gases (the difference between the scattering cross sections when the signs of the amount ξ of circular polarization of the incident and scattered photons change simultaneously)[8] and explain, in particular, the results of the experiments of Weihe *et al.*[7] The importance of allowing for dissipation effects in elliptically polarized fields follows from simple qualitative ideas. In view of the irreversibility of dissipation processes, allowing for dissipation introduces into the problem T-odd (changing sign under time reversal) parameters Γ characterizing the intensity of such processes. On the other hand, polarization anomalies in an elliptically polarized field can be caused only if the problem contains the axial T-odd vector $\xi\mathbf{k}$ (with $\mathbf{k}$ the wave vector), which is characteristic only of an elliptically polarized wave. As a result, when we allow for dissipation, the cross sections or other physical characteristics of the problem may contain "dichroic" terms proportional to $\xi\Gamma$ and changing sign when $-\xi$ is substituted for ξ, while in nondissipative media these effects are forbidden by, say, general space–time symmetry restrictions.

2. THE POLARIZATION PARAMETERS OF HARMONICS

Let $\mathbf{e}$ and $\boldsymbol{\kappa}$ be the unit complex-valued polarization vector and the unit vector along the direction of propagation of the pump field,

$$\mathbf{F}(\mathbf{r},t)=2F\ \mathrm{Re}\{\mathbf{e}\exp[i(\mathbf{kr}-\omega t)]\},\quad \mathbf{ee}^*=1,$$

with $\boldsymbol{\kappa}=\mathbf{k}/|\mathbf{k}|$, and $\mathbf{e}\boldsymbol{\kappa}=0$. To analyze polarization effects in multiphoton processes it is convenient to employ the following invariant (with respect to the choice of the coordinate system) parametrization of $\mathbf{e}$ for the general case of arbitrary polarization of the light field $\mathbf{F}(\mathbf{r},t)$ with an ellipticity parameter γ:

$$\mathbf{e}=\frac{\boldsymbol{\epsilon}+i\gamma[\boldsymbol{\kappa},\boldsymbol{\epsilon}]}{\sqrt{1+\gamma^2}},\quad -1\leqslant\gamma\leqslant 1, \tag{1}$$

where $\boldsymbol{\epsilon}$ is the unit vector along the principal axis of the polarization ellipse of $\mathbf{F}(\mathbf{r},t)$, which, obviously, coincides with $\mathbf{e}$ in the case of linear polarization, and

$$l=\frac{1-\gamma^2}{1+\gamma^2}=\mathbf{ee}=\mathbf{e}^*\mathbf{e}^*,\quad \xi=\frac{2\gamma}{1+\gamma^2}=i\boldsymbol{\kappa}[\mathbf{ee}^*] \tag{2}$$

are the degrees of linear and circular polarization, with ξ coinciding with the standard Stokes parameter ξ_2 (see Ref. 9), and $l^2+\xi^2=1$ for totally polarized radiation.

We examine the interaction of the field $\mathbf{F}(\mathbf{r},t)$ with a gas of freely oriented atoms or molecules in the electric dipole approximation. The state of an atom is described by a state vector $|0\rangle=|\gamma_0 J_0 M_0\rangle$, where J_0 and M_0 are the total angular momentum and its projection on the quantization axis, and γ_0 stands for the energy E_0 and the other quantum number of the atomic level. The amplitude $A^{(n\omega)}(F,\mathbf{e},\mathbf{e}')$ for generation of a harmonic with frequency $\nu\omega$ and polarization $\mathbf{e}'$ measured by a detector is proportional to the amplitude of the coherent merging of n photons of the strong field and can be written in terms of the atomic polarization vector at frequency $n\omega$ averaged over M_0:

$$A^{n\omega}(F,\mathbf{e}',\mathbf{e})\propto\mathbf{e}'^*\mathbf{P}_{n\omega},\quad \mathbf{P}(t)=2\,\mathrm{Re}\{\mathbf{P}_{n\omega}\exp(-in\omega t)\}. \tag{3}$$

With these definitions, the qualitative features of the polarization effects in a strong elliptically polarized fields follow from fairly general considerations.

In an arbitrary perturbation-theory order N ($N>n$) the expression for $\mathbf{P}_{n\omega}^{(N)}$ contains n $\mathbf{e}$ vectors (according to the number of absorbed photons needed for generating a harmonic at frequency $n\omega$) and $(N-n)/2$ pairs of vectors $\mathbf{e}$ and $\mathbf{e}^*$ corresponding to processes of "reemission" of photons, out of which pairs the polar vector $\mathbf{P}_{n\omega}^{(N)}$ is formed. Since an even number of vectors $\mathbf{e}$ and $\mathbf{e}^*$ cannot form a polar vector, only odd harmonics can be generated, with generation being absent when the pump radiation is circularly polarized, since in this case $\mathbf{ee}=\mathbf{e}^*\mathbf{e}^*=0$. These well-known facts follow directly from the selection rules for dipole radiation when nonlinear susceptibilities are calculated by perturbation-theory techniques, but remain valid in ultrastrong fields, where the common nonlinear-susceptibility formalism breaks down. They also remain valid if we allow for nondipole effects in the atom–field interaction:[10] although in this case the problem acquires an additional vector $\mathbf{k}$ and the polarization vector is nonzero even for even values of n, the polarization vector is collinear to $\mathbf{k}$, with the result that harmonic generation (zero-angle scattering) is impossible.

It is convenient to represent the polarization structure of $\mathbf{P}_{n\omega}$ in the general case of arbitrary intensity of the pump field as the sum of the contribution $\mathbf{P}_{n\omega}^{(0)}$ of the first nonvanishing perturbation-theory order and the "nonlinear" part $\mathbf{P}_{n\omega}^{(NL)}$ (we use the atomic system of units):

$$\mathbf{P}_{n\omega}=\mathbf{P}_{n\omega}^{(0)}+\mathbf{P}_{n\omega}^{(NL)}=l^{(n-1)/2}F^n\{\chi^{(0)}(-n\omega)\mathbf{e}+F^2[(\chi_\parallel-\chi_\perp)\mathbf{e}+l\chi_\perp\mathbf{e}^*]\}. \tag{4}$$

Here $\chi^{(0)}(-\nu\omega)\equiv\chi^{(n+1)}_{xx\ldots x}(-n\omega;\omega,\ldots,\omega)$ is the component of the ordinary nonlinear-susceptibility tensor of rank $n+1$ for $|\gamma_0 J_0 M_0\rangle$, which after averaging over M_0 is left with only one linearly independent component, and $\chi_\parallel$ and $\chi_\perp$ are generalized susceptibilities, which depend not only on ω but also on the parameters F and l of the strong field. Both $\chi_\parallel$ and $\chi_\perp$ are defined in such a way that, as $F\to 0$, they become the two linearly independent components of a tensor of rank $n+3$ determining the lowest-order perturbation-theory correction ($\sim F^2$) to $\xi^{(0)}(-n\omega)$ (see Ref. 6).

In the region where the perturbation-theory series in F converge, the susceptibilities $\chi_\parallel$ and $\chi_\perp$ have the following general structure:

$$\chi_{\parallel,\perp}(F^2,l^2,\omega)=\sum_{k=0}^{\infty}\left(\sum_{p=0}^{k}\chi_{\parallel,\perp}^{(n+2k+3,p)}(\omega)l^{2p}\right)F^{2k}. \tag{5}$$

Here the $2k+1$ coefficients $\chi_{\parallel,\perp}^{(n+2k+3,p)}$ with $p=0,1,\ldots,k$ for a fixed k can be expressed in terms of $2k+2$ linearly independent components of the susceptibility tensor of rank $n+2k+3$ determining a correction $\sim F^{2k+2}$ to $\chi^{(0)}(-n\omega)$. As is known,[11] perturbation-theory series in a monochromatic field have a finite radius of convergence in F, which for nonresonant frequencies $\omega<|E_0|$ yields $F\leqslant F_{cr}\propto\omega^{3/2}$. Thus, at frequencies ω of order $0.1|E_0|$, typical of experiments in harmonic generation in strong fields, one should expect that perturbation-theory estimates of $\chi_\parallel$ and $\chi_\perp$ are meaningful only for intensities $I\leqslant 10^4\ \mathrm{W\ cm^{-2}}$.

Equation (4) shows that the polarization of the harmonics corresponds to the polarization $\mathbf{e}$ of the pump radiation only in the case of linear polarization or in a weak field, where the dependence of a harmonic's intensity on the degree of linear polarization l of the pump radiation has a simple power-like form:[6]

$$I_{n\omega}(F,\mathbf{e})\propto l^{n-1}F^{2n}|\chi^{(0)}(-n\omega)|^2. \tag{6}$$

The situation is quite different in the case of a strong field with a nonzero degree of circular polarization. The susceptibilities

$$\chi_1\equiv\chi^{(0)}(-n\omega)+F^2(\chi_\parallel-\chi_\perp),\quad \chi_2\equiv F^2\chi_\perp$$

in Eq. (4) generally have an imaginary (skew-Hermitian) part, which is related to the probability of an atom being ionized in a strong field and serves in the present case as the dissipation process discussed earlier. The imaginary part of $\chi^{(0)}(-n\omega)$ is nonzero only for $n\geqslant N_0$, where $N_0=[|E_0|]+1$ is the minimum number of photons needed, according to the law of energy conservation, to ionize the atom (here we ignore the resonant case, where the real occupation of the resonant level is the dissipation process and Im $\chi^{(0)}$ is proportional to the width of the resonant level). In a strong field the imaginary parts of $\chi_{\parallel}$ and $\chi_{\perp}$ are nonzero at all values of ω, since even in the perturbation-theory limit and with $n \ll N_0$ the terms in the series (5), starting from $k=N_0-n$, acquire an imaginary part. Thus, strictly speaking, the approximation of a transparent gaseous medium is inapplicable for describing harmonic generation in a strong light field.

Because of the non-Hermitian nature of $\chi^{(0)}$, $\chi_{\parallel}$, and $\chi_{\perp}$ in a strong field, not only does the ellipticity $\gamma^{(n\omega)}(F)$ of a harmonic's polarization differs from the value for the pump field and depend on intensity, but so does the orientation of the ellipse axes.

Defining the axis ratio and the orientation of a harmonic's polarization ellipse in terms of the complex-valued amplitude $\mathbf{P}_{n\omega}$ [Eq. (4)] in the standard way,[9] we arrive at the following expression for the degree of circular polarization of a harmonic:

$$\xi^{(n\omega)}(F)=\frac{2\gamma^{(n\omega)}}{1+(\gamma^{(n\omega)})^2}=\xi\frac{|\chi_1|^2-l^2|\chi_2|^2}{|\chi_1+l\chi_2|^2}. \qquad (7)$$

The fact that $\xi^{(n\omega)}$ is proportional to ξ follows from general ideas. Allowing only for the principal terms in the perturbation-theory expansion (5) for χ_1 and χ_2 and assuming $|\chi_{\parallel,\perp}^{(n+3,0)}|F^2 \ll |\chi^{(0)}(-n\omega)|$, we can easily see that in a weak field the relative variation of $\gamma^{(n\omega)}$ is proportional to the pump field intensity,

$$\frac{\gamma^{(n\omega)}(F)-\gamma}{\gamma}=-2lF^2\frac{\mathrm{Re}[\chi^{(0)}(-n\omega)\chi_{\perp}^{(n+3,0)}]}{|\chi^{(0)}(-n\omega)|^2}, \qquad (8)$$

and follows from the results of Ref. 6 if we ignore dissipation.

The angle θ of the principal axis rotation of a harmonic's polarization ellipse in relation to $\boldsymbol{\epsilon}$ is determined by

$$\tan 2\theta=\frac{2\xi\ \mathrm{Im}(\chi_1^*\chi_2)}{|\chi_1|^2+l^2|\chi_2|^2+2\,\mathrm{Re}(\chi_1^*\chi_2)}. \qquad (9)$$

We see that θ increases monotonically with ξ, which fully agrees with the experimental data.[7] Allowing only for the first two nonvanishing perturbation-theory orders for χ_1 and χ_2, we arrive at an expression similar to (8):

$$\tan 2\theta=\frac{2\xi F^2}{|\chi^{(0)}(-n\omega)|^2}[\mathrm{Re}\ \chi^{(0)}(-n\omega)\mathrm{Im}\ \chi_{\perp}^{(n+3,0)} - \mathrm{Im}\ \chi^{(0)}(-n\omega)\mathrm{Re}\ \chi_{\perp}^{(n+3,0)}]. \qquad (10)$$

The sign of θ depends on the ratio of the real and imaginary parts of $\chi_{1,2}$. In the experiment described in Ref. 7 the signs of θ are different for harmonics with $n \leq 9$ and $n \geq 11$, while the absolute value of θ for the same values of γ changes little. This fact, not emphasized by Weihe *et al.*,[7] appears to be very important because it points to the rapid variation in the ratio of the real and imaginary parts of the generalized susceptibilities at $n \sim N_0$ ($N_0=10$ for argon). In particular, the acute "sensitivity" of the imaginary parts of χ_1 and χ_2, which are related to the ionization probability, to the number of absorbed photons can serve as an indication that in the optical frequency range the tunneling ionization mechanism does not occur in pure form even in the case of strong fields.

In the case of perturbation theory, the reason why the sign of θ changes at the n-photon ionization threshold follows directly from (10). For $n<N_0$ we have Im $\chi^{(0)} \times(-n\omega)=0$, and the sign of θ is determined by the sign of the ratio Im $\chi_{\perp}^{(n+3,0)}\chi^{(0)}$. For $n>N_0$, $\chi^{(0)}(-n\omega)$ acquires an imaginary part, and the change in the sign in θ means that

$$\frac{\mathrm{Im}\ \chi^{(0)}}{\mathrm{Re}\ \chi^{(0)}}>\frac{\mathrm{Im}\ \chi_{\perp}^{(n+3,0)}}{\mathrm{Re}\ \chi_{\perp}^{(n+3,0)}}. \qquad (11)$$

Note that in their experiment on measuring the dependence of the harmonic yield on the parameter γ in Ne and He in the field of a Ti:sapphire laser with $I \sim 10^{15}$ W cm^{-2}, Burnett *et al.*[5] also observed anomalies (the nonmonotonic dependence on γ) for $n \sim N_0$, which, according to what has been said above, are also caused by dissipation effects related to the threshold of the N_0-photon ionization channel. These anomalies are the manifestation of the well-known threshold singularities in the energy dependence of the reaction cross sections for potentials with a Coulomb asymptotic behavior:[12] when a new reaction channel (allowed by the energy conservation law) opens, the cross section experiences a jump caused by the sudden appearance of an imaginary part in the reaction amplitude and then monotonically grows with energy up to the threshold of the next channel. At present there are no data on the dispersion dependence and ratio of the real and imaginary parts of the atomic susceptibilities in the strong-field limit. Numerical calculations of $\chi^{(0)}(-3\omega)$ for hydrogen in a broad frequency range have been carried out with perturbation-theory techniques.[13] The results obtained in Ref. 13 show that at the three-photon ionization threshold ($\omega=\omega_t \equiv |E_{1s}|/3$) the imaginary part of $\chi^{(0)}(-3\omega)$, equal to zero for $\omega<\omega_t$, exceeds the real part by a factor of almost 100. Apparently, for the higher-order susceptibilities $\chi_{\perp}^{(n+3,0)}$ the difference between the real and imaginary parts in the near-threshold region is not so large, which leads to the inequality (11) and a qualitative explanation of the results of Ref. 7.

Note that the pronounced threshold anomalies must flatten out for short-range potentials, since in this case the susceptibilities experience no jumps at the threshold frequencies $\omega_n=|E_0|/n$, and the imaginary parts, which emerge when a new ionization channel is opened, have only order-2 branch-point threshold singularities and vary monotonically, starting from zero at the threshold. This situation can be implemented in experiments with negative ions. In this connection it must be noted that the calculations of harmonic generation done with the model of delta-function potentials mentioned in Ref. 7 can hardly provide quantitative agreement with atomic experiments.

3. ELLIPTIC DICHROISM IN STRONG-FIELD HARMONIC GENERATION

Measurements of the polarization of a harmonic as such and of the total harmonic yield

$$I_{n\omega}(F,\mathbf{e})\propto l^{n-1}F^{2n}\{|\chi_1|^2+l^2[|\chi_2|^2+2\,\mathrm{Re}(\chi_1\chi_2^*)]\} \tag{12}$$

do not allow complete data to be extracted on the quantitative relationship between the real and imaginary parts of χ_1 and χ_2. Detailed information can be obtained by measuring the intensity $I_{n\omega}(F,\mathbf{e},\mathbf{e}')$ of a harmonic component polarized in a certain way (with a polarization vector $\mathbf{e}'$); this component can be extracted by a polarization analyzer and is given by the square of the absolute value of the amplitude in (3).

Combining (1) and (2) and allowing for a similar parametrization of $\mathbf{e}'$ by γ', ξ', and l' (here we have $\boldsymbol{\kappa}'=\boldsymbol{\kappa}$ according to the requirement that the wave vectors of the pump field and the harmonic must be collinear), for the most general case of arbitrary polarizations (including partial polarizations) of the pump field and the detector we arrive at the expression

$$\begin{aligned} I_{n\omega}(F,\mathbf{e},\mathbf{e}')\propto l^{n-1}F^{2n}[&|\chi_1|^2(1+ll'\cos 2\phi+\xi\xi') \\ &+l^2|\chi_2|^2(1+ll'\cos 2\phi-\xi\xi') \\ &+2l\,\mathrm{Re}(\chi_1\chi_2^*)(l+l'\cos 2\phi) \\ &+2\xi ll'\,\mathrm{Im}(\chi_1\chi_2^*)\sin 2\phi], \end{aligned} \tag{13}$$

where ϕ is the angle between the principal axes of the polarization ellipses (the vectors $\boldsymbol{\epsilon}$ and $\boldsymbol{\epsilon}'$) of the pump field and the detected polarization $\mathbf{e}'$. In deriving (13) we employed the following relationships:

$$\begin{aligned} &(\mathbf{ea})(\mathbf{e}^*\mathbf{b})=\mathrm{Re}\{(\mathbf{ea})(\mathbf{e}^*\mathbf{b})\}-\frac{i}{2}\,\xi\,\mathbf{k}[\mathbf{ab}], \\ &2\,\mathrm{Re}\{(\mathbf{ea})(\mathbf{e}^*\mathbf{b})\}=2l(\mathbf{a}\boldsymbol{\epsilon})(\boldsymbol{\epsilon}\mathbf{b})+(l-1)[\mathbf{ak}][\mathbf{kb}], \end{aligned} \tag{14}$$

which are valid for real vectors $\mathbf{a}$ and $\mathbf{b}$ and follow from Eqs. (1) and (2). We see that polarization measurements for different geometries make it possible to independently measure all four atomic parameters in (13) containing two complex-valued generalized susceptibilities and in this way to carry out a complete experiment. In weak fields the main contribution is provided by the first term on the right-hand side of Eq. (13), while measuring the other parameters makes it possible to extract information about the corrections to the first nonvanishing perturbation-theory order and, correspondingly, to establish the limits of applicability of the theory in describing generalized susceptibilities.

The last term on the right-hand side of Eq. (13) is the most interesting. It originates from the vector combination

$$\mathrm{Im}\{(\mathbf{ee}')(\mathbf{ee}'^*)\}=\xi l'(\boldsymbol{\epsilon\epsilon}')(\boldsymbol{\kappa}[\boldsymbol{\epsilon\epsilon}'])=\frac{1}{2}\,\xi l'\sin 2\phi \tag{15}$$

and leads to dichroism, i.e., the difference in the generation cross sections that emerges when the signs of the amount of circular polarization of all the photons change simultaneously. For instance, all other things being equal, the yield of the linearly polarized component of a harmonic ($l'=1$) depends on the sign of the degree of polarization of the pump field, i.e., on the sense of rotation of the vector $\mathbf{F}(\mathbf{r},t)$. Such left–right asymmetry is caused by a combination of dissipation and strong-field effects, since it is the presence of T-odd dissipation parameters in the problem that leads to a nonzero coefficient of the vector combination (15) containing a polar T-odd vector $\boldsymbol{\kappa}$. At the same time, in a weak field (in the first nonvanishing perturbation-theory order) the presence of a skew-Hermitian part in the susceptibility $\chi^{(0)}\times(-n\omega)$ leads to no polarization anomalies. In contrast to circular dichroism in the scattering of light[8] or in bremsstrahlung and electron–atom scattering in a light field,[14,15] where the effect is the strongest at $\xi=\pm1$, in multiphoton processes the magnitude of dichroism is determined by the product ξl and is nonzero only when polarization is elliptic ("elliptic dichroism"). One manifestation of elliptic dichroism, the sharp difference between the angular distribution of photoelectrons in multiphoton ionization of atoms in an elliptic field and the distribution in the case of linear or circular polarization, was observed for the first time in the experiments of Bashkansky *et al.*[16]

Thus, by using elliptically polarized pump radiation we can extract information about nonlinear susceptibilities inaccessible in experiments with purely linear or circular photon polarization. Polarization measurements with $n\sim N_0$ are especially interesting becuase of the possibility of using the results of measurements in analyzing the applicability of various theoretical approaches to describe the process of harmonic generation and ionization of atoms in a strong light field. We also note in conclusion that in experimental measurements of elliptic dichroism it may be simpler to measure the difference in harmonic yield for two orthogonal linear polarizations $\mathbf{e}'$ with $\phi=\pm\pi/4$, which is equivalent to substituting $-\xi$ for ξ, as Eq. (13) implies.

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