Parametric up-conversion of infrared radiation and its applications

É. S. Voronin and V. L. Strizhevskiĭ

M. V. Lomonosov State University, Moscow
and T. G. Shevchenko State University, Kiev
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The results of theoretical and experimental investigations of conversion of infrared radiation to the visible (or ultraviolet and near infrared) range are summarized. An analysis is made of the physical basis of the process, which involves generation of the sum or difference frequency in media with a quadratic nonlinear polarizability. In particular, the critical and noncritical phase matching, the angular and spectral widths of the phase-matching curve, the energy and quantum efficiency of the conversion process, and similar topics are considered. The applications of the conversion process in detection of infrared signals, infrared vision, and nonlinear infrared spectroscopy are dealt with. Typical results obtained so far are given briefly. The following topics are discussed: the criteria for selection of nonlinear media useful for the near and middle infrared range; detection of thermal infrared signals; response time and other parameters of nonlinear media; methods for the calculation of the spatial (angular) structure of the output radiation and its relationship to the structure of the initial infrared field; spatial resolution and number of resolved elements in parametric infrared viewers; comparison of systems based on tangential (noncritical) and critical vector phase matching; and so on. Spectral resolution and response time of nonlinear infrared spectroscopic methods are discussed. It is shown that various parametric detector analyzers for the infrared range are in many respects not inferior or considerably better than conventional infrared devices. This applies particularly to the response time because parametric converters have no competitors in, for example, picosecond infrared spectroscopy. Extensive use of parametric infrared converters in science and technology will represent a new stage in the mastering of the infrared range.

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CONTENTS

1. INTRODUCTION

Parametric up-conversion occupies a special place among the great variety of nonlinear optical phenomena and processes.[1-3] The importance of this phenomenon is primarily due to its use for conversion of infrared signals to the visible (less frequently, to the ultraviolet) range, which is frequently called "visualization" of infrared radiation. The phenomenon is very interesting from the physical point of view and it can be used to detect and analyze infrared radiation by the well-developed methods available for the visible range.

The fact that a converted signal retains basically the information carried by the original infrared signal is of fundamental importance. This information may be carried by the angular structure or by the frequency spectrum. Therefore, practical applications of parametric up-conversion of infrared signals include not only infrared signal detection but also development of infrared viewers, and of analyzers of infrared spectra (nonlinear infrared spectrometers). Another important application is infrared holography.[4,5]

The use of up-conversion is particularly important in the middle infrared range near 10 μ. This range corresponds to a "window" of atmospheric transparency; moreover, the emission frequency of the most powerful

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infrared coherent source (CO_2 laser) and maximum intensity of black body radiation at room temperature, etc. lie in this range. The existing infrared detectors and analyzers suffer—in spite of the considerable progress made recently[6,7]—from a number of shortcomings and on the whole they are considerably inferior to detectors of visible radiation: their sensitivity, temporal and spectral resolution, and the number of resolved elements are all too low; strong cooling is required as well as screening from the background, etc. True, heterodyne detectors[7,8, etc.] are exceptional in respect of sensitivity but their use requires a highly stable local oscillator, coherent radiation, etc. The heterodyne detection method is used mainly under laboratory conditions for detection of CO_2 laser radiation and it does not provide a universal solution to the problem of detection.

Studies of parametric conversion of infrared radiation have started right from the beginning of the present stage of development of nonlinear optics. Already in 1962 Armstrong *et al.*[9] pointed out the desirability of detection of infrared radiation by up-conversion because of the low noise level characterizing this process. Fairly systematic investigations started beginning from 1967-1968. The number of papers continued to grow in subsequent years with the result that the physical laws governing this phenomenon are now well known and the favorable prospects of practical applications have become clearly apparent. In view of this, it would be desirable to summarize the results obtained so far, which will be our main purpose. Several reviews have been published on this subject[10-13] but they do not reflect the current status and have dealt mainly with some particular aspects of the subject, particularly with visualization of infrared images.

2. PHYSICAL BASIS OF CONVERSION

a) Generation of sum and difference frequencies in parametric up-conversion of infrared radiation

Parametric up-conversion of infrared radiation involves mixing of an infrared signal with suitable auxiliary radiation (pump) in a nonlinear medium; this produces new radiation at the sum or difference frequency, which carries the information in the original infrared signal. The basic idea of this conversion process is illustrated in Fig. 1. A nonlinear crystal receives a pump wave ω_1 and an infrared signal ω_2; the output is in the form of a wave $\omega_3 = \omega_1 + \omega_2$ or $\omega_3 = \omega_1 - \omega_2$ (the incident waves $\omega_{1,2}$ are then filtered out).[1)]

The physical cause of mixing of the waves $\omega_{1,2}$ is an

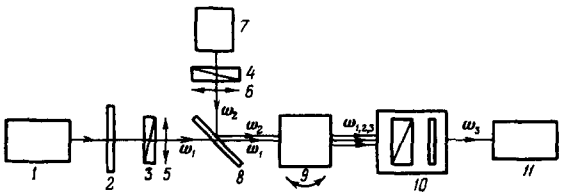

FIG. 1. Block diagram of a system for parametric up-conversion of infrared radiation: 1) pump laser; 2) background filter; 3), 4) polarizers; 5), 6) beam-shaping lenses; 7) infrared source; 8) semitransparent plate; 9) nonlinear crystal; 10) filter unit; 11) detection unit.

electric nonlinearity, which is the nonlinear coupling between the specific polarization of the medium P and the electric field E. The processes of interest to us are described by quadratic nonlinear polarization[1-3] $P_i^{NL} = \chi_{ijk} E_j E_k$ (repeated indices of tensors will always denote summation). Here, $\chi_{ijk}(\omega_1, \omega_2)$ is the quadratic nonlinear polarizability, which is a third-rank tensor; information on the properties of this tensor can be found in Refs. 1–3. In the transparency range it is usually assumed that $\chi_{ijk}(\omega_1, \omega_2) = \chi_{ijk}(\omega_1, \omega_1) = 2\chi_{ijk}^{2\omega_1}$, where $\chi_{ijk}^{2\omega_1}$ is the tensor responsible for second harmonic generation, on which there is fairly complete information.[14-16, etc.] The tensor $\chi_{ijk}^{2\omega_1}$ is symmetric with respect to the second and third indices and, therefore, it is convenient[17] to describe it by a matrix d_{im} replacing the index pair (j, k) by the index m in accordance with the rule: $11 \to 1, 22 \to 2, 33 \to 3, 23(32) \to 4, 13(31) \to 5, 12(21) \to 6$.

Usually only waves with fixed linear polarizations, which can be described by the unit vectors $\mathbf{e}_{1,2,3}$ interact efficiently. The only important component[1] is $P^{NL} = (\mathbf{e}_3, \mathbf{P}^{NL})$, and in the case of interaction between plane monochromatic waves

$$\mathbf{E}_\nu(\mathbf{r}, t) = \mathbf{e}_\nu A_\nu e^{i(k_\nu r - \omega_\nu t)} + \text{c.c.},$$
$$k_\nu = \frac{\omega_\nu}{c} n_\nu = \frac{2\pi}{\lambda_\nu} n_\nu, \quad \nu = 1, 2, 3$$

it can be represented in the form

$$\left.\begin{aligned} P^{NL} &= P_0 e^{i(\mathbf{q}\mathbf{r} - \omega_3 t)} + \text{c.c.}, \quad P_0 = \chi A_1 A_2, \\ \mathbf{q} &= \mathbf{k}_1 \pm \mathbf{k}_2, \quad \chi = 2d = e_{3i} e_{1j} e_{2k} \chi_{ijk}(\omega_1, \omega_2). \end{aligned}\right\} \tag{1}$$

The scalar quantity χ (or d) is known as the effective nonlinear polarizability. Calculations of this quantity for various specific cases can be found, for example, in the book by Zernike and Midwinter.[3] An infrared wave is usually much weaker than a pump wave. Therefore, the latter can almost always be regarded as having a constant intensity.

In specific illustrations of the theory we shall consider only the interaction of waves in uniaxial crystals. We shall use "*o*" and "*e*" to denote ordinary and extraordinary waves. Interactions for various combinations of polarizations (different types of interaction) will be denoted by $oo \to e$, $oe \to e$, etc., where the three letters will represent in turn the polarizations of the three waves $\omega_{1,2,3}$, respectively. A crystal will be regarded as a plane-parallel layer of thickness l, whose faces are perpendicular to the z axis.

[1)] We shall throughout use the indices 1, 2, and 3 for the quantities referring to the pump wave, infrared radiation and sum or difference frequency waves, respectively. The majority of the general results for the sum and difference frequencies are basically similar (with the exception of the sensitivity to infrared signals and structure of the wavefront); to be specific, we shall usually consider sum frequency generation. We shall assume that a nonlinear crystalline medium is nonmagnetic, weakly anisotropic, and transparent at all three frequencies $\omega_{1,2,3}$.

b) Phase matching and various ways of its realization

Generation of ω_3 is most effective if the wave ω_3 which reaches a given element of volume from other elements is in phase with the radiation created in this element. This favorable phase relationship is obtained if

$$\mathbf{k}_3 = \mathbf{q} = \mathbf{k}_1 \pm \mathbf{k}_2. \qquad (2)$$

The condition (2) is known as the phase or wave matching.[1-3] It may be satisfied for various wave polarizations in anisotropic crystals along certain directions of their propagation when the dispersion of the refractive index is compensated by the anisotropy. Under phase-matching conditions usually employed in practice the intensity of the output radiation is enhanced by several orders of magnitude because of the cumulative process along the whole crystal.

Phase matching can be conveniently illustrated by means of wave-vector surfaces.[18,19] Figure 2 shows sections of the surface formed by the XZ plane (the Z axis is parallel to the optic axis c). We shall consider a negative uniaxial crystal (this type of crystal is most important in practice) and the $\omega_3 = \omega_1 + \omega_2$ process with the $oo \rightarrow e$ interaction. We shall first construct the whole surface of the wave vector $\mathbf{k}_1$ (which is a sphere) and select some specific direction of this vector. Taking the end of the vector $\mathbf{k}_1$ as the origin of the new coordinate system with axes parallel to the initial ones, we now plot the surface of the vector $\mathbf{k}_2$ (again a (sphere). Finally, in the initial coordinate system we plot the surface $\mathbf{k}_3$ (ellipsoid of revolution).

Depending on the angle θ_1 we can have various situations. As long as this angle is small (Fig. 2a), we have $k_3 > |\mathbf{k}_1 + \mathbf{k}_2|$ for any direction of $\mathbf{k}_2$ because of the normal dispersion in the transparency region ($n_{30} > n_{1,20}$). However, because of bending of the surface $\mathbf{k}_3$ in the case of sufficiently large values of θ_1, the surface $\mathbf{k}_2$ and $\mathbf{k}_3$ come into contact (Fig. 2b), naturally if the degree of anisotropy is sufficiently high. At the point of contact the vector triangle $\mathbf{k}_{1,2,3}$ closes and the condition (2) is satisfied. A further increase in θ_1 alters the contact (tangency) to an intersection at two points A and B (in space—along a certain closed line), as shown in Fig. 2c. Therefore, the case of Fig. 2c is known as the critical vector phase matching, whereas the case of Fig. 2b is known as the noncritical (tangential) phase matching. The latter was discovered[2)] in 1968 by Warner.[20,21] The tangential phase matching in the oo

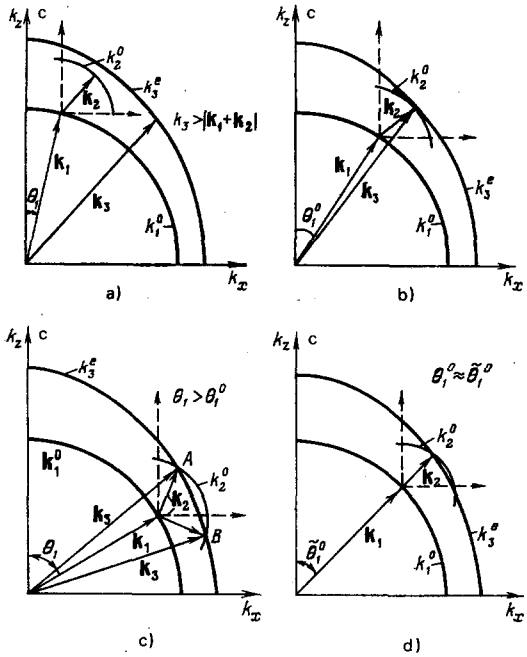

FIG. 2. Relative positions of wave-vector surfaces at frequencies $\omega_{1,2,3}$ ($oo \rightarrow e$ interaction): a) no phase matching for any $\mathbf{k}_2$; b) tangential phase matching; c) critical vector phase matching; d) one-dimensional critical phase matching. Here, c is the optic axis of the crystal.

$\rightarrow e$ interaction is always slightly vectorial (because of anisotropy). Collinear (one-dimensional) critical phase matching (Fig. 2d) is close to the last case.

The importance of the tangential phase matching in parametric conversion of infrared images is due to the considerable increase in the angular width of the field of vision of the infrared signal. Warner[20] as well as Voronin *et al.*[25,26] specified and analyzed the geometric conditions for such conversion. A crystal should be cut so that trangential phase matching occurs when the pump radiation travels along the normal to the faces (z axis). In practice, selection of the cut results only in coarse matching, whereas precision phase matching is obtained by a slight inclination relative to the z axis.[25] It is convenient to begin with attainment of one-dimensional phase matching by rotating a crystal around the y axis at right-angles to $\mathbf{k}_1$ and then to rotate it additionally about the same axis through a known angle φ_0 governed by the anisotropy. The direction $\mathbf{k}_2$ corresponding to tangential phase matching then makes a known angle ψ_0 with $\mathbf{k}_1$. The values of the angles ψ_0 and φ_c are listed in Table I. It is essential to know in advance, at least approximately, the required orientation of the vector $\mathbf{k}_1$. The relevant information on the phase-matching angles θ_1^0 and $\tilde{\theta}_1^0$ (Fig. 2) can be found, in particular, in Refs. 11 and 27 and also in Refs. 28 and 29. The situation is optimal when the tangential phase matching is obtained for 90° (or close to this angle).

If the phase-matching condition is not obeyed, i.e., if there is a phase mismatch $\tau = \mathbf{k}_3 - \mathbf{k}_1 - \mathbf{k}_2$, the intensity of the output radiation decreases rapidly as the mismatch increases. In the case of the transversely homogeneous conversion process with constant plane waves $\omega_{1,2}$ we have $\tau \parallel \mathbf{z}$ and the dependence of the intensity of

[2)]The idea of tangential phase matching was first formulated in 1968 by Midwinter[21] with a reference to a personal communication from Warner. Warner himself published the relevant results[20] in 1969. The existence of noncritical phase matching was demonstrated even earlier[22] in second harmonic generation with collinear pump and harmonic wave vectors, both perpendicular to the optic axis; the usefulness of this 90° geometry lies in the absence of aperture effects[23,24] etc resulting from a transverse drift of energy because of the difference between the directions of the phase and group wave velocities. It should be pointed out that the phase matching "critical" in the X, Z plane is noncritical in the perpendicular plane. The phase matching in the case discussed by Warner[20] is noncritical in any plane.

TABLE I. Angular characteristics of tangential phase matching.

Sum frequency generation $\omega_3 = \omega_1 + \omega_2$			Difference frequency generation $\omega_3 = \omega_1 - \omega_2$		
Type of interaction	ψ_0	φ_0	Type of interaction	ψ_0	φ_0
$oo \to e$	$\frac{k_3}{k_1}\gamma_3$	$\frac{k_2}{2k_1}\gamma_3$	$eo \to o$	0	0
$eo \to e$	$\frac{k_2}{k_1}\gamma_3$	$\frac{k_2 k_3 \gamma_3^2}{2k_1(k_3\gamma_3 - k_1\gamma_1)}$	$eo \to e$	$\frac{k_3}{k_1}\gamma_3$	$\frac{k_2 k_3 \gamma_3^2}{2k_1(k_3\gamma_3 - k_1\gamma_1)}$
$oe \to e$	$\frac{k_3}{k_1}(\gamma_3 - \gamma_2)$	$\frac{k_2 k_3 (\gamma_3 - \gamma_2)^2}{2k_1(k_3\gamma_3 - k_2\gamma_2)}$	$ee \to o$	$-\frac{k_3}{k_1}\gamma_2$	$\frac{k_2 k_3 \gamma_3^2}{2k_1(k_3\gamma_3 - k_1\gamma_1)}$

*This table was compiled by N. E. Kornienko. The notation is as follows: ψ_0 and φ_0 are the angles formed by the vector $\mathbf{k}_2$ in tangential phase matching and by the direction of collinear phase matching, respectively, with the vector $\mathbf{k}_1$ corresponding to tangential phase matching. The angles are assumed to be positive if measured in the direction toward the optic axis; γ is the anisotropy angle obeying $\tan\gamma = \varepsilon\sin 2\theta/2(1+\varepsilon\sin^2\theta)$, where $\varepsilon = (\varepsilon_\perp - \varepsilon_\parallel)/\varepsilon_\perp$ and $\theta = (\mathbf{k}, \mathbf{c})$.

the output radiation on the phase mismatch is given by the factor obtained in Sec. 2c $\zeta(x) = \sin c^2 x = (\sin x/x)^2$, where $x = \tau_z l/2$; this factor differs considerably from zero in the $|x| \leq \pi/2$ range, i.e., $|\tau_z| \leq \pi/l$. This imposes restrictions on the permissible angular and spectral mismatch (detuning) for each of the waves. These can be represented by the effective angular ($\psi_{1,2}$) and spectral ($\delta_{1,2}$) widths of the phase-matching curve. They correspond to doubled values of the angular and frequency mismatch[3)] corresponding to $|x| = \pi/2$. The linear approximation for τ_z expressed in terms of the angular and frequency mismatch corresponds to the critical phase matching. If $\tau_z = 0$ in the linear approximation, we have to include second-order terms; this corresponds to noncritical phase matching. In parametric conversion of infrared images it is necessary to ensure noncritical angular phase matching of the infrared signal.

We shall now consider some important special cases.

An analysis of the one-dimensional critical phase matching can be found in Ref. 28. Some idea of the values of ψ_2^{cr} can be obtained from Fig. 3 (based on Ref. 28). For the $oo \to e$ interaction, we have $\psi_1^{cr} = (k_2/k_1)\psi_2^{cr}$, i.e., the requirements in respect of the divergence of the pump radiation are more stringent ($\psi_1^{cr} < \psi_2^{cr}$). An analysis of the case of basically vector critical phase matching is given in Ref. 30. In this case the values of $\psi_{1,2}^{cr}$ are approximately an order of magnitude smaller. According to Ref. 25, in the noncritical (tangential) phase matching case we have $\psi_2^{nc} = 2\sqrt{2\pi\mu k_3/k_1 k_2 l}$, and for $\omega_1 \gg \omega_2$, we obtain $\psi_2^{nc} = 2\sqrt{\mu\lambda_2/n_2 l} \sim \sqrt{\lambda_2/l}$. For $\lambda_2 = 10\mu$, $l = 1$ cm, and $n_2 = 2$, we find that $\psi_2^{nc} \approx 2°35'$. Usually ψ_2^{nc} is considerably greater than $\psi_2^{cr\,(oo\to e)}$, but in the one-dimensional $oe \to e$ case the difference between ψ_2^{cr} and ψ_2^{nc} decreases or disappears completely.

[3)]Sometimes the width of the phase-matching curve is deduced from the condition $|x| = \mu\pi/2$, assuming that $\mu = 0.885$, which corresponds to $\zeta(x) = 1/2$; here, $\mu = 2$ corresponds to the first zeros of $\zeta(x)$, etc. All the numerical data are given in the present paper for $\mu = 1$.

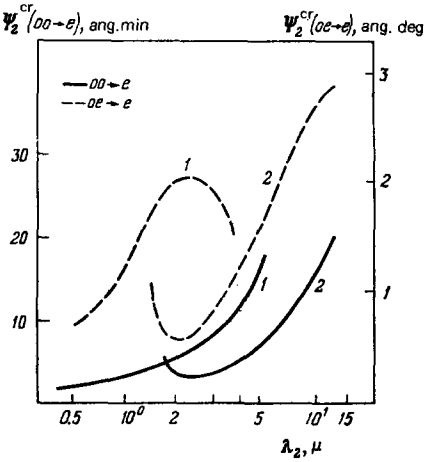

FIG. 3. Angular widths of one-dimensional critical phase-matching curves $\lambda_1 = 0.6943\ \mu$:[28] 1) $LiIO_3$; 2) Ag_3AsS_3.

Karpenko *et al.*[28] also analyzed the spectral widths of the phase-matching curve. Figure 4, taken from Ref. 28, gives the dependence $\delta_2(\lambda_2)$ for a number of variants of the collinear interaction. We can see that δ_2 varies within the range ~1–100 cm^{-1}. For selected values of λ_2 we may also attain "group" (noncritical in respect of the frequency ω_2) phase matching; the value of δ_2 then increases by about two orders of magnitude.[31] Finally, in some special cases we can have phase matching which is noncritical in respect both of the angle and of the frequency.[31]

c) Conversion efficiency

A mathematical description of the interaction of the $\omega_{1,2,3}$ waves due to the nonlinearity of the polarization

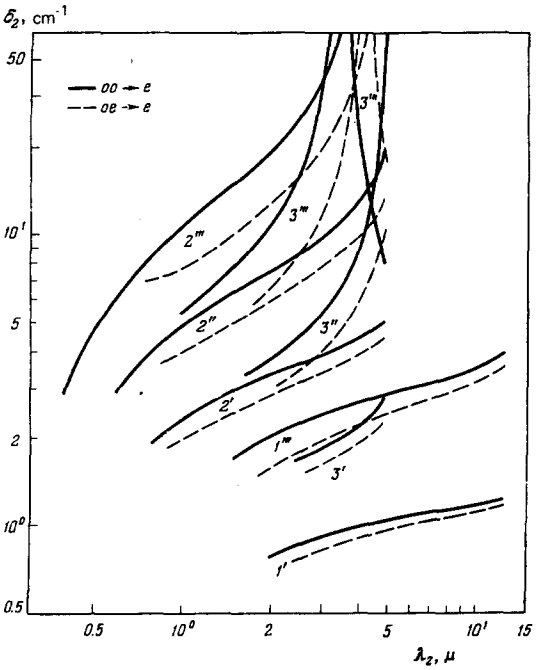

FIG. 4. Dependence of the spectral width of the phase-matching curve δ_2 on λ_2: 1) Ag_3AsS_3; 2) $LiIO_3$; 3) $LiNbO_3$; single prime denotes $\lambda_1 = 0.488\ \mu$, two primes denote $\lambda_1 = 0.6943\ \mu$, and three primes denote $\lambda_1 = 1.06\ \mu$.

P^{NL} of Eq. (1) is based on the use of the familiar method of truncated equations.[1,2] In the steady-state transversely homogeneous approximation the system of truncated equations for coupled scalar amplitudes of the field is[1-3]

$$\left.\begin{aligned}
&\frac{dA_1}{dz}=iQ_1A_3A_2^*e^{i\tau_z z}, \quad \tau_z=k_{3z}-k_{1z}-k_{2z}, \quad \omega_3=\omega_1+\omega_2,\\
&\frac{dA_2}{dz}=iQ_2A_3A_1^*e^{i\tau_z z}, \quad Q_\nu=\frac{2\pi\omega_\nu}{cn_\nu\cos\xi_\nu\Lambda_\nu},\\
&\frac{dA_3}{dz}=iQ_3A_1A_2e^{-i\tau_z z} \quad (\nu=1,\ 2,\ 3), \quad \Lambda_\nu=1-\frac{e_{\nu z}(\mathbf{k}_\nu\mathbf{e}_\nu)}{k_{\nu z}}\approx 1;
\end{aligned}\right\} \tag{3}$$

here, ξ_ν are the angles between $\mathbf{k}_\nu$ and the z axis. The system (3) should be supplemented by the boundary conditions $A_{1,2}|_{z=0}=A_{1,2}^0, A_3|_{z=0}=0$. The problem has a general solution which is rather cumbersome and contains elliptic functions.[1,2] However, in those cases when the fields $\omega_{1,2}$ can be regarded as constant, only the third equation of the system remains and this is solved easily. Going over directly to the intensities $I_\nu=cn_\nu|A_\nu|^2/2\pi$ and assuming that $I_3\sim I_1I_2$, we introduce the energy $\eta_e=I_3/I_2^0$ and quantum $\eta_q=N_3/N_2^0$ conversion efficiencies[4] ($N_\nu=I_\nu/\hbar\omega_\nu$ is the flux density of photons of frequency ω_ν):

$$\eta_q=\frac{\omega_2}{\omega_3}\,\eta_e=aI_1l^2\operatorname{sinc}^2\frac{\tau_z z}{2}, \quad a=\frac{32\pi^3\omega_2\omega_3d^2}{c^3n_1n_2n_3\cos^2\xi_3}; \tag{4}$$

here, $I_1=P_1/S$ and P_1 is the energy flux of the pump radiation in an interaction zone whose transverse cross section is S. The constant a represents the quantum efficiency of parametric conversion in a crystal of unit thickness for unit pump intensity and exact phase matching.

If a constant infrared field is not assumed, then

$$\eta_q=\frac{\sin^2(l\sqrt{aI_1+\tau_z^2/4})}{1+(\tau_z^2/4aI_1)}. \tag{5}$$

We can see that η_q oscillates with l and that $\eta_q^{\max}=[1+\tau_z^2/4aI_1)]^{-1}$; in the case of phase-matched generation we have $\eta_q^{\max}=1$. Thus, the range of values of η_q is limited to the interval $[0,1]$. Physically, this is due to the fact that an elementary event underlying the process of the sum frequency generation is the "coalescence" of two photons $\hbar\omega_1$ and $\hbar\omega_2$ producing a third photon $\hbar\omega_3$, so that the number of new photons $\hbar\omega_3$ is exactly equal to the loss of the infrared photons $\hbar\omega_2$. Naturally, $N_3^{\max}$ cannot exceed N_2^0. On the other hand, $\eta_e=(\omega_3/\omega_2)\eta_q>\eta_q$, since $\hbar\omega_3>\hbar\omega_2$ and, if ω_1 is sufficiently high, η_e can obviously exceed 1.

In the case of difference frequency generation, we find that Eq. (5) is modified to

$$\eta_q=\left|\operatorname{ch}f+\frac{i\tau_z}{2f}\operatorname{sh}f\right|^2, \quad f=\sqrt{aI_1-\frac{\tau_z^2}{4}}.$$

An elementary event now represents "dissociation" of a photon $\hbar\omega_1$ into two photons: $\hbar\omega_1\to\hbar\omega_2+\hbar\omega_3$; the energies of both waves $\omega_{2,3}$ are at the same time increased. Therefore, not only η_e but also η_q can, in principle, exceed 1 by any amount.

It is important to note that in all cases the conversion efficiency is independent of the intensity of the infrared input signal and is governed only by the pump intensity. In practice, focused Gaussian beams are frequently used; this case is discussed theoretically in Ref. 32. The results of Ref. 32 are generalized in Refs. 33–38 to the case of elliptic focusing, allowing for diffraction and aperture effects; this is done for the collinear[33,34] and perpendicular (transverse)[35-38] interactions. The optimal focusing ensures[34] an increase in η_q for proustite ($\lambda_2=10.6\mu$) by a factor of ~30; for $P_1=1W$ and 1 cm, we can obtain $\eta_q\sim10^{-4}$.

3. DETECTION OF INFRARED SIGNALS BY PARAMETRIC UP-CONVERSION

a) Materials employed

The most important nonlinear media with a nonzero quadratic nonlinear polarizability are noncentrosymmetric crystals.[1,2] They can be used also for parametric conversion of infrared radiation. For this purpose, they must satisfy the following conditions: 1) the nonlinearity should be as high as possible; 2) phase matching should be attainable for any type of interaction between the waves; 3) they should be transparent at frequencies of all three waves $\omega_{1,2,3}$; 4) the optical quality (homogeneity, absence of impurities, etc.) should be high and samples of sufficiently large dimensions should be available; 5) optical strength in the presence of various reversible and irreversible changes of the parameters should be high for operation at high pulse or average pump powers; 6) mechanical machining should be easy, crystals should not be hygroscopic, etc. Additional requirements may have to be satisfied, depending on the application and conditions of use.

A fairly large group of well-tried crystals is now available and these satisfy quite well the above conditions or most of them. Information on these crystals

TABLE II. Nonlinear crystals usually employed in parametric conversion from near infrared.

Crystal	Crystallographic class *) and optical sign	Transparency range, μ	Nonlinear polarizability, 10^{-9} cgs esu	Wavelength μ	References
KH_2PO_4 (KDP) (potassium dihydrogen phosphate)	D_{2d} ($\bar{4}2m$),	0.2—1.5	$d_{36}=3.0\pm0.6$	**)	4, 5, 11, 25, 29, 39–47
$NH_4H_2PO_4$ (ADP) (ammonium dihydrogen phosphate)	D_{2d} ($\bar{4}2m$), —	0.2—1.5	$d_{36}=3.4\pm0.7$	**)	58–60
$LiNbO_3$ (lithium niobate)	C_{3v} ($3m$), —	0.4—5.0	$d_{22}=20\pm7$ $d_{31}=36\pm13$ $d_{33}=160\pm160$	1.06	11, 22, 28, 31, 44, 57, 61–65
$LiIO_3$ (lithium iodate)	C_6 (6), —	0.3—5.5	$d_{31}=38\pm10$ $d_{33}=34\pm11$	1.06	28, 42, 57, 84, 86–105
$Ba_2NaNb_5O_{15}$ (barium-sodium niobate)	C_{2v} ($mm2$), Biaxial crystal	0.4—4.0	$d_{31}=100\pm30$	1.0	11, 106

*The class is given in the Schonflies notation; the international symbol is given in parentheses.[17]

**The value of d_{36} depends weakly on the wave-length in the range $1-0.6\mu$.

[4] Here, the index 0 of I_2 and N_2 denotes the value at the entry to the crystal (in the $z=0$ plane). In the case of a given infrared field, both I_2 and N_2 are independent of z but the definitions of the quantities η_e and η_q remain valid also when conversion of an infrared signal is quite considerable.

TABLE III. Nonlinear crystals usually employed in parametric conversion from near and middle infrared, including $\lambda_2 \sim 10\ \mu$ range.

Crystal	Crystallographic class and optical sign	Transparency range, μ	Nonlinear polarizability, 10^{-9} cgs esu	Wavelength μ	References
Ag_3AsS_3 (proustite)	C_{3v} $(3m)$, —	0.64—13	$d_{15}=80$ $d_{22}=128\pm8$ $d_{31}=80$	1.5 1.5	11, 20, 27, 33–36, 66, 80–82, 95, 107–140
$AgGaS_2$ (silver thiogallate)	D_{2d} $(\bar{4}2m)$, —	0.49—12.5	$d_{14}=270\pm176$ $d_{14}=86\pm39$ $d_{14}=138$	10.6 10.6 1.06	13, 82, 83 141–143
$ZnGeP_2$ — (zinc-germanium phosphide)	D_{2d} $(\bar{4}2m)$, +	0.74—12	$d_{14}=540\pm240$	10.6	12, 107, 117, 142, 144, 145
GaSe (gallium selenide)	D_{3h} $(\bar{6}2m)$, —	0.65—18	$d_{22}=380$	10.6	13, 136, 139, 146, 147
HgS (cinnabar)	D_3 (32), +	0.6—13.5	$d_{11}=240\pm80$ $d_{11}=300\pm105$	10.6	11, 13, 107, 145, 148

TABLE IV. Optical breakdown thresholds I_{th} for giant pulses applied to some nonlinear crystals used in up-conversion of infrared signal frequencies ($\lambda_1 = 1.06\ \mu$).

Crystal	KDP, ADP	$LiNbO_3$	$LiIO_3$	$Ba_2NaNb_5O_{15}$
I_{th} MW/cm²	500—600 [162, 163]	1000 [162, 163]	100 [162, 163]	10—100 [162, 163]

Crystal	Ag_3AsS_3	$ZnGeP_2$	GaSe	$AgGaS_2$
I_{th} MW/cm²	20—25 *), **) [12, 165, 166]	4 ***) [167]	30 [146]	20 ****) [164]

*According to Ref. 164, surface damage appears in Ag_3AsS_3 when the intensity is a few megawatts per square centimeter.
**No breakdown is observed even after 1000 pulses of 12.5 MW/cm² intensity.[12,165]
***According to Ref. 142, $ZnGeP_2$ is damaged by 5–10 pulses of $I = 20$ MW/cm² intensity but a single pulse of $I = 50$ MW/cm² intensity is sufficient for damage. This crystal can withstand cw CO_2 laser intensities of ~ 10 W/cm².
****Samples grown recently in the Soviet Union[141,168] have a higher optical strength: the damage threshold of 100–200 nsec duration is 30 ± 3 MW/cm² (Ref. 143).

is given in Tables II and III. We can see that in the near infrared range the most frequently used crystals are those of KDP, $LiNbO_3$, and $LiIO_3$. In the middle infrared range the most widely used crystal is proustite, but one should also mention the very promising (as found recently) $AgGaS_2$ and GaSe crystals. Nonlinear polarizabilities are listed for the generation of a second harmonic $2\omega_1$; the absolute values of the polarizabilities are given.

This does not exhaust the list of crystals employed by various workers. We can add pyrargyrite Ag_3SbS_3 (Refs. 148–150), $LiGaO_2$ (Ref. 151), CsH_2AsO_4 (CDA) and RbH_2AsO_4 (RDA) (Ref. 53), α-HIO_3 (Ref. 11), sodium nitrite $NaNO_2$ (Ref. 152), rubidium chlorate $RbClO_3$ (Ref. 153), molecular cyrstal of metanitroaniline (Refs. 154 and 155), etc. Naturally, other nonlinear crystals can also be used. Reutov and Tarashchenko[156] visualized He–Ne laser radiation ($\lambda_2 = 1.152\mu$, $\lambda_1 = 1.06\mu$) in an optical microwaveguide with a carrier layer of lithium niobate.[156] Finally, there have been several investigations of the possibility of using gaseous resonance media, particularly alkali metal vapors[157–161] and other materials.

An important limiting factor is the optical strength of crystals. The threshold power densities I_{th} corresponding to optical breakdown by giant pulses are listed in Table IV. In the cw regime the process of optical damage naturally occurs at much lower power densities (intensities). For example, in the case of proustite the average threshold values of the power densities are ~100 W/cm² (Refs. 12, 27, 118, 164, and 165). On the other hand, according to Ref. 141, $AgGaS_2$ crystals show no signs of optical breakdown at power densities of ~1 kW/cm² and according to Ref. 143, this is true even at ~100 kW/cm².

The material parameters of a crystal which determine the efficiency of parametric conversion of infrared signals occur in Eq. (9) as the factor $x = d^2/n^3$ (n is the average refractive index). Therefore, the factor x is frequently called the quality parameter representing the efficiency of a crystal.[146] The values of x for some crystals (in units of 10^8 cgs esu) are as follows[146] for $\lambda_1 = 1.06\mu$, $\lambda_2 = 10.6\mu$, and $oo-e$ interactions: 2.3 for $ZnGeP_2$, 1.7 for GaSe, 0.23 for Ag_3AsS_3, and 0.17 for $AgGaS_2$. These values—together with the data in Tables III and IV—demonstrate the high promise of GaSe crystals.

One should mention particularly $AgGaS_2$ (silver thiogallate) crystals. Their importance and promise have been greatly enhanced by the synthesis[168] of highly transparent (with the absorption coefficient less than 0.1 cm^{-1} in the 0.6–12μ range) samples. It would be very desirable to prepare also $ZnGeP_2$ crystals with sufficiently high transparency, which has not been done so far.

b) Brief review of main results

Systematic investigations of parametric up-conversion of infrared signals started approximately in 1967. An analysis of the earlier results was given in Refs. 10–12 and we shall not consider them in detail except for some special aspects. Investigations have been carried out in the near and middle infrared range; in the case of the far infrared range one can mention only the interesting work of Takatsuji[169] who considered theoretically the detection of $\lambda_2 > 50\mu$ radiation in quartz, CdS, and ZnTe crystals; he estimated the sensitivity limit as $\sim 10^{-5}$–10^{-6} W for $I_1 \sim 10^5$ W/cm² and he found that the main noise source was the laser radiation background. Parametric conversion of far infrared radiation has not yet been given sufficient attention.

Right from the beginning, attempts have been made to increase the conversion efficiency and thus the sensitivity of the method. Already Midwinter and Warner[64] achieved in their pioneering work a quantum efficiency $\eta_q \sim 1\%$ for conversion of mercury lamp radiation ($\lambda_2 = 1.65 - 1.9\mu$, $I_2 \sim 1\mu$W/cm²) with giant ruby laser pulses in $LiNbO_3$; the sensitivity of this process was $\sim 10^{-8}$W/cm² and it was limited by infrared impurity luminescence. The cw conversion efficiency was much lower but already in Ref. 66 a value of $\sim 10^{-11}$W ($P_1 = 0.1$W, $LiNbO_3$) was reported.

Radiation of the 10.6μ wavelength was first converted to the visible range by Warner[110] in proustite ($\eta_q = 1.4 \times 10^{-6}$). Although other crystals have been used for this purpose, proustite has remained without serious competition up to 1975 because large samples of high optical quality can be grown. New possibilities have appeared since the preparation in the Soviet Union[168] of high-transparency samples of silver thiogallate[83,141-143] and of gallium selenide.[146,147] The advantages of the thiogallate $AgGaS_2$ are its high optical strength and relatively weak anisotropy, which permits optimal focusing of the radiation, weak temperature dependence of the refractive index, and the ability of conversion to shorter visible wavelengths, together with the nonlinearity close to that of proustite, etc. The available GaSe samples are also highly transparent, have a large quality parameter x, and make it possible to penetrate further into the infrared range. However, it is difficult to machine GaSe samples. The search for new crystals is continuing.

The sensitivity of a parametric infrared signal detector is governed primarily by the conversion noise. In the case of difference frequency generation the noise is governed by the usual parametric scattering.[170] In the case of sum frequency generation, one may expect also second-order parametric noise.[59,171] An exhaustive analysis of noise was carried out by Il'inskii and Petnikova,[172] who showed that the second-order parametric noise is important in the near infrared (according to Ustinov *et al.*[98] this noise is suppressed in the range $\omega_1 < \omega_2$), whereas in the middle infrared range the thermal and background noise components are important. Il'inskii and Petnikova also determined the photocount statistics of the converted radiation,[173,174] particularly the influence of pump fluctuations[175,176] and stray light reaching a photodetector.[177,178] Boyd and Kleinman[32] analyzed in detail the noise in a compund system comprising a noisy infrared radiation converter and a photodetector for recording the converted radiation. In particular, they derived an important formula for the noise equivalent power (NEP).

Experimental investigations of the spontaneous radiation noise in sum frequency generation are reported in Refs. 57, 179, and 180. The noise due to impurity luminescence is considered in Refs. 64, 78, and 181; the noise associated with fluctuations of the pump radiation in the case of insufficient filtration is considered in Ref. 66, etc. Thermal and background noise in the vicinity of 10μ was investigated in detail by Lucy *et al.*[27,120,125] Suppression by cooling a crystal made it possible to record thermal radiation emitted by a plate held at $T = -30$°C (Ref. 120). An experimental investigation was also made[101] of the photocount distribution for the dark photomultiplier current and converted signal ($\lambda_2 = 1.15\mu$, $\lambda_1 = 1.06\mu$, $LiNbO_3$) and an infrared signal of 5×10^{-14}W power was detected. The sensitivity achieved has been higher than for other infrared detectors, with the exception of the heterodyne systems.

A detailed analysis of the experimental aspects of increasing the up-conversion efficiency[10,12,27,123] has been carried out with special attention to the 10μ range and systems based on proustite.[12,27] One should distinguish detection of reflected radiation signals in the case of illumination of objects by coherent light and detection of thermal radiation emitted by the objects themselves.

Detection of thermal radiation in the 6.5–12.5μ range at temperatures near the room value was first achieved by Falk and Yarborough.[114] The thermal contrast, limited primarily by pump fluctuations, was a few degrees. Lucy[27] analyzed thermal source radiation (a "gray" body with $T = 600$°C) in proustite in an interval 0.5μ wide near 10.6μ.

Wide-band near-infrared radiation has been made visible in several investigations. A system of critical vector phase matching with strong focusing of the $\omega_{1,2}$ beams[52,88,89] has been found useful for the purpose. Radiation emitted by a Nernst rod[62,76] and by a Globar[61,63,70,74,75,78,79] has been frequently converted in $LiNbO_3$.

The contrast of an object is much better when it is illuminated (for example, with CO_2 laser radiation) and the reflected radiation is observed. Milton[12] analyzed the relevant applied aspects and reached the conclusion that devices of this type designed for special applications are highly competitive (in contrast to the case of infrared viewing, for which η is much less) with, for example, well-known infrared scanning devices.[13]

The principal way of increasing the parametric conversion efficiency is to increase the pump radiation intensity (power density) and to use crystals with high degree of nonlinearity. For example, Krivoshchekov, Marennikov, *et al.*[92,93] used $P_1 = 15$ MW and achieved $\eta_q = 12.5$–15% ($\lambda_1 = \lambda_2 = 1.06\mu$) in $LiIO_3$. In the cw regime Smith and Mahr[69] achieved $\eta_q = 10^{-4}$ ($\lambda_1 = 0.5145\mu$, $\lambda_2 = 2.5$–4.5μ) in $LiNbO_3$.

The use of pulse-periodic pump pulses and particularly the employment of pulse-periodic pump and infrared pulses synchronized at frequencies $\omega_{1,2}$ has been an important step forward. This has made it possible to use relatively high peak powers and thus increase η_q without damaging a nonlinear crystal.[115,116,123] For example, the value $\eta_q \approx 2 \times 10^{-3}$ has been obtained[123] for $\lambda_2 = 10.6\mu$ employing a pulse repetition frequency $f = 133$ Hz. This efficiency is a record value for conversion in the 10.6μ range outside the resonator, with the exception of pumping with ultrashort pulses for which the efficiency $\eta_q \approx 0.1$ has been obtained.[129]

The next important step in increasing theparametric conversion efficiency has been the placing of a nonlinear crystal in a laser resonator.[53,85,91,99,101,108,122,143,182] In this way Gurskii[91] first reached the efficiency limit $\eta_q = 1$ ($LiIO_3$, $\lambda_2 = 3.39\mu$). Moreover, he converted thermal radiation of 3.4μ wavelength to the visible range inside a resonator. Such internal (intracavity) conversion of thermal and laser radiation from the near-infrared range was also achieved by others.[99,85,101,183] Voronin, Solomatin, and Shuvalov[108,122] were the first to achieve intracavity conversion of $\lambda_2 = 10.6\mu$ radiation in proustite and to increase the efficiency by a factor of ~20 ($\eta_q = 1.5 \times 10^{-3}$). A similar system based on an $AgGaS_2$ crystal[143] made it possible to reach $\eta_q = 1.6 \times 10^{-3}$ in the

cw and 0.4 in the pulse ($\tau = 0.5$ μsec) regimes. Kornienko, Ryzhkov, and Strizhevskii[184] studied theoretically the intracavity up-conversion of infrared radiation under conditions when the conversion process affected considerably the operation of a master laser. Akhmanov and Dmitriev[185] proposed the use of difference frequency generation in an optical resonator by a traveling pump wave using pulses of duration less than the time required to establish stable oscillations ("parametric superregenerator"). In this case the difference frequency output should be proportional to the initial infrared signal and the energy conversion efficiency η_e can reach ~10^4. Such a device should, in spite of the relatively high noise level, be very effective for investigating the kinetics of many fast infrared processes.

In the search for the optimal solution, Voronin *et al.*[37,38] (see also Ref. 140) investigated a system with mutually perpendicular pump and infrared beams ($\lambda_2 = 10.6\mu$, proustite). This ensured spatial decoupling of the $\omega_{1,2}$ beams, which made it easier to suppress the pump background, facilitated the use of antireflection coatings on the opposite faces operating separately on the $\omega_{1,2}$ waves, simplified realization of intracavity conversion,[108] etc. Another successful original solution was double conversion of $\lambda_2 = 10.6\mu$ radiation by pumping with a YAG:Nd^{3+} laser:[83] the first stage was an $AgGaS_2$ crystal with $\eta_q = 0.3$ producing $\lambda_3 = 0.97\mu$ and the second stage was an $LiIO_3$ crystal with $\eta_q = 0.8$ producing $\lambda_3 = 0.507\mu$ (this corresponded to a high sensitivity range of photocathodes) or α-HIO_3 (see also Ref. 142). In this way it was possible to achieve in the 10μ range the level of sensitivity attainable in photomultipliers sensitive to visible radiation. Il'inskii and Petnikova[82] discussed theoretical aspects of such conversion.

Another promising possibility is associated with the use of gaseous resonance media.[157-161] Such media are centrosymmetric and, therefore, parametric conversion can only be achieved because of a cubic (or higher order) nonlinearity. The anomalous rise of this nonlinearity under resonance conditions in a suitable gas can make this a very strong effect. For example, Harris *et al.*[160] employed two-photon resonance and achieved infrared radiation conversion in sodium vapor from about 10μ to a range near 3320 Å with an efficiency η_q up to 58%, which corresponded (because of the high value of ω_3) to $\eta_e = 1620\%$.

TABLE V. Summary of typical efficiencies of *cw* parametric up-conversion of infrared signals.

λ_2, μ	λ_1, μ	P_1, W	Sum (s) or difference (d) frequency	λ_3, μ	Nonlinear crystal	η_q	η_e	References
2.5—4.5 (cosmic source)	0.5145	3	s	0.45	$LiNbO_3$	10^{-4}	$8\cdot10^{-4}$	69
5.2—6.4	0.6328	$6.6\cdot10^{-3}$	d	0.7	Ag_3AsS_3	$5\cdot10^{-5}$	$4.2\cdot10^{-4}$	137
10.6	0.647	$6\cdot10^{-2}$	d	0.69	Ag_3AsS_3	10^{-7}	$1.5\cdot10^{-6}$	35
10.6	0.6764	$9.8\cdot10^{-4}$	s	0.635	Ag_3AsS_3	$6.6\cdot10^{-9}$	$1.1\cdot10^{-7}$	128
2.4—3.4 (Nernst rod)	0.488	0.1	s	0.42	$LiNbO_3$	$2.5\cdot10^{-4}$	$1.6\cdot10^{-3}$	63
10.6	1.15	$2\cdot10^{-3}$	s	1.04	GaSe	$1.2\cdot10^{-6}$	$1.2\cdot10^{-5}$	138

TABLE VI. Summary of typical efficiencies of parametric conversion of infrared signal by sum frequency generation resulting from *Q*-switched laser pumping.

λ_2, μ	λ_1, μ	P_1 or I_1	λ_3, μ	Nonlinear crystal	η_q	η_e	References
1.7 (mercury lamp)	0.6943	$I_1 = 2$ MW/cm²	0.49	$LiNbO_3$	10^{-2}	$3.5\cdot10^{-2}$	64
0.7 (xenon lamp)	1.06	$I_1 = 100$ MW/cm²	0.42	KDP	$2\cdot10^{-2}$	$4\cdot10^{-2}$	48, 49
1.06	0.6943	$P_1 = 50$ MW	0.42	KDP	$2\cdot10^{-3}$	$5\cdot10^{-3}$	50, 52
1.06	1.06	$P_1 = 8.5$ MW	0.53	$LiIO_3$	0.125	0.25	92
1.6—3 (Globar)	0.6943	$I_1 \sim 3$ MW/cm²	0.54	$LiNbO_3$	1	4.3	78
10.6	0.6943	Ultrashort pulses	0.65	Ag_3AsS_3	0.107	1.75	129
9.26 CO_2	0.53	$I_1 = 50$ MW/cm²	0.3305	Sodium vapor	0.58	16.2	157, 160

Tables V–VIII summarize typical experimental values of the efficiency of parametric up-conversion of infrared signals. We can see that in present-day experiments it is possible to achieve $\eta_q \sim 10^{-4}$–10^{-6} in the cw regime and ~1–10^{-1} in the *Q*-switched pulse regime. The values $\eta_q \sim 1$–10^{-1} are obtained also when a nonlinear crystal is placed inside a laser resonator (cavity). In the pulse-periodic regime the quantum efficiency is $\eta_q \sim 10^{-2}$–10^{-3} for the individual pulses. In many cases $\eta_e > 1$ can be achieved. A very striking case is that of gases for which $\eta_e = 16.2$ can be obtained (Table VI). These values of η are sufficient for many applications.

The results reported in the present subsection do not exhaust all the aspects of parametric up-conversion of infrared signals as a means for their detection. One should mention also investigations of the influence of random inhomogeneities in a crystal on the value of η (Ref. 186); an analysis of the conditions for recording the radiation of frequency ω_3 with slow-response image amplifiers in the pulse regime;[94] investigations of the dependence $\eta_q(I_1)$ under multimode pumping conditions when saturation of η_q at 50% (in contrast to the harmonic pumping case) is observed;[102] application of the method under discussion for recording and analysis of spectral[108,113,133,135] and temporal[129,140,187] structures of CO_2 laser radiation and also of infrared radiation generated in various physical processes (see, for example, theoretical studies of Raman scattering in the infrared

TABLE VII. Summary of typical efficiencies of parametric conversion of middle-infrared radiation by sum frequency generation resulting from pulse-periodic pumping with YAG:Nd^{3+} laser radiation ($\lambda_1 = 1.064$ μ, $\lambda_2 = 10.6$ μ, $\lambda_3 = 0.967$ μ).

Power per pulse P_1, W	Average power P_1, W	Pulse duration τ, μsec	Pulse repetition frequency f, Hz	Nonlinear crystal	η_q	η_e	References
740	0.002	0.02	133	Ag_3AsS_3	$8\cdot10^{-4}$	$8.4\cdot10^{-3}$	116
$1.8\cdot10^{-3}$	0.005	0.02	133	Ag_3AsS_3	$2\cdot10^{-3}$	$2\cdot10^{-2}$	123
190	0.8	0.7	$6\cdot10^{-3}$	$AgGaS_2$	$3.6\cdot10^{-2}$	0.4	141
100	1	0.01	100	GaSe	$2\cdot10^{-4}$	$2.2\cdot10^{-3}$	146
10^4	0.02	0.1	20	$AgGaS_2 + LiIO_3$ *)	0.24	5	83

*Double parametric conversion, first in $AgGaS_2$ and then in $LiIO_3$, generating radiation with $\lambda_3 = 0.507$ μ.

TABLE VIII. Summary of typical efficiencies of parametric up-conversion of infrared radiation by sum frequency generation in nonlinear crystals placed inside pump laser cavity.

λ_2, μ	λ_1, μ	Average laser power P_1, W	λ_3, μ	Nonlinear crystal	η_q	η_e	References
3.39 (He-Ne) and 3.4 (thermal radiation)	0.6943	1500	0.58	$LiIO_3$	1	6	91
10.6	1.064	15	0.967	Ag_3AsS_3	$1.5 \cdot 10^{-3}$	$1.5 \cdot 10^{-2}$	108, 122
1.15 (He—Ne)	1.064	1500	0.55	$LiIO_3$	0.38	0.8	101
1.064	1.064		0.532	$LiIO_3$	0.125	0.25	105
10.6	1.064	1500	0.967	$AgGaS_2$	0.4	4.8	143

range[188] as well as studies of parametric oscillation[103] etc.), applications in extending the tuning range of parametric oscillation[103] etc.), applications in extending the tuning range of parametric optical oscillators,[85,104,182] precision measurements of the velocity of light in vacuum,[135] and many others.

Some of these important applications will be considered in greater detail later in the present review.

c) Some experimental aspects

Figure 1 is a block diagram of a typical system for the experimental realization of parametric up-conversion of infrared signals. It is suitable for the collinear interaction of the $\omega_{1,2}$ waves (obvious changes can be made for the perpendicular propagation of these two waves). Figure 5 shows schematically an intracavity parametric conversion system used in Refs. 108 and 122. It is important to note that most of the essential and many of the auxiliary procedures are standard; this applies to, for example, focusing, alignment of a crystal to the phase-matching orientation, filtering of the output radiation, etc. Shaping of infrared beams requires special (for example, germanium) optics. In the case of relatively low values of η_q there is a considerable likelihood that a photodetector receives stray laser pump radiation. Careful filtering of the output radiation is then needed. In the $\mathbf{e}_1 \perp \mathbf{e}_2$ case one can use particularly the polarization filtering. However, this may be insufficient. In those cases when $\omega_2 \ll \omega_1$ and $\omega_3 \approx \omega_1$, interference filters have to be employed. This, of course, weakens to some extent the useful signal and such weakening needs to be allowed for specially. For example, it is reported in Ref. 27 ($\lambda_1 = 0.6943\mu$, $\lambda_2 = 10.6\mu$) that the pump radiation was attenuated by a factor of 10^{16} by a Glan polarizer and six filters ranged in series; the transmission of the wave at the sum wavelength of $\lambda_3 = 0.6516\mu$ was 15%. The purity of the spatial and spectral structure of the pump radiation also plays an important role.

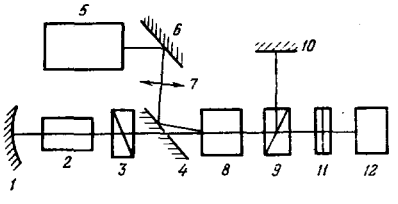

FIG. 5. Block diagram of a system for parametric conversion of infrared radiation in a laser cavity (resonator):[108,122] 1), 4), 6), 10) mirrors; 2) YAG :Nd^{3+} active element; 3), 9) polarizers; 5) CO_2 laser; 7) lens for shaping infrared beam; 8) nonlinear crystal (proustite); 11) filter; 12) recording unit.

d) Characteristics of parametric up-conversion of thermal radiation

The principal distinguishing feature of thermal radiation is its wide-band nature. Therefore, for this reason (because the spectral width of the phase-matching curve δ_2 is considerable) the efficiency of parametric conversion of collimated thermal radiation is relatively low. In such cases it is convenient to have a large value of δ_2. A change in the direction of propagation of infrared radiation in a crystal results in conversion in a different part of the spectrum because the phase-matching angle is a function of the frequency. It follows that the field of view in the observation of thermal sources is wider. Similarly, the width of the converted-radiation spectrum $\Delta\omega_2$ increases with the angle of reception. These aspects are considered in Refs. 12, 27, 114, 177, etc.

Radiation from hot bodies appears against the background of thermal radiation of the ambient medium (atmosphere). The latter fluctuates continuously. Therefore, there is a certain smallest temperature contrast ΔT which can be detected. According to Refs. 12, 81, and 177, this contrast is

$$\Delta T = \frac{A}{e\Omega\psi_3\sqrt{P_1\tau}}, \quad A = \frac{1.13}{dB/dT}\sqrt{\frac{\hbar\omega_2 B}{\delta_2 \varkappa_3 a}},$$

where Ω is the solid angle of reception of the infrared radiation; τ is the time needed for integration in detection of the converted radiation; ψ_3 is the angle of reception of the converted radiation (in radians); $\varkappa_3$ is the quantum efficiency of the detector at the frequency ω_3; a is the reduced parametric conversion frequency of Eq. (4). For $\tau \sim 1$ sec and $P_1 \sim 1$ W, it is quite realistic to expect detection of a contrast amounting to $\Delta T \sim 1$°K or less. However, this requires a high transverse homogeneity (to within at least 0.2%) of the pump radiation and homogeneity of the photocathode surface to within at least 1% (Ref. 117), which is not easy to achieve in practice.

e) Sensitivity of parametric infrared detectors. Comparison with other detectors

We shall end this section by considering the sensitivity of a parametric infrared radiation detector, which consists of a parametric converter and a detector of the converted radiation (usually a photodetector). An important relative characteristic of the photodetector sensitivity[6] is the noise equivalent power (NEP). The product $\mathrm{NEP} \cdot \sqrt{\Delta f}$ (Δf is the effective band width of the photodetector) determines the minimum detectable signal.

Boyd and Kleinman[32] analyzed the NEP of the parametric detector and derived the following expression:

$$\mathrm{NEP} = \frac{1}{\eta_{ic}}\sqrt{(\mathrm{NEP}_3)^2 + \frac{2\hbar\omega_3}{\varkappa_3} P_{3n}},$$

where NEP_3 refers to a photodetector at the frequency ω_3; P_{3n} is the noise power at the frequency ω_3. It is

usually possible to ignore the term P_{3n} in the near infrared range[62,69,91,119,149] and then we have NEP $=\eta_e^{-1}$NEP$_3$ (except in those cases when $\eta_e \gtrsim 0.1$—Ref. 189).

Table IX gives typical values of the NEP. We can see that the best values ~10^{-14}–10^{-15} W·Hz$^{-1/2}$ are obtained in the near infrared range. In the 10μ range the NEP is somewhat poorer (10^{-10} W·Hz$^{-1/2}$), which is related primarily to an increase in the role of thermal and background noise, and to filtering difficulties. However, theoretical calculations[128] suggest that this can be improved to ~10^{-13} W·Hz$^{-1/2}$.

We should also bear in mind that the response time of parametric detectors is governed only by the time constant of the visible-radiation detector, which is usually 10^{-9} sec or shorter, or by the reciprocal of the pass-band width, which is $\lesssim 10^{-10}$ sec. Consequently, in some cases corresponding to $\delta_2 \sim 10$–100 cm^{-1} the up-conversion process has no competition[140] in the case of ultrashort infrared pulses ($\tau < 10^{-10}$ sec). It follows from all this discussion that parametric near-infrared detectors are now not inferior—in respect of the NEP and response times—to the best of the tranditional direct detectors and some types of parametric detector are considerably better. In contrast to direct detectors, good NEP and response can be attained at the same time. The advantages of parametric detectors in the middle-infrared range ($\lambda_2 = 10\mu$) can be realized only if further work is carried out. Clearly, the attainable NEP is of the same order of magnitude as in the case of the best cooled bolometers, but the time constant is considerably shorter.

In comparing parametric and heterodyne detectors[7,8] it is desirable to use the absolute sensitivity Q, because the two types of detector operate under different noise conditions: the parametric detectors are subject to a thermal or quantum noise of the visible radiation detector and the heterodyne detectors suffer from the "noise-in-the-signal." If we take NEP $= 10^{-14}$ W·Hz$^{-1/2}$ in the former case and the sensitivity per unit frequency interval 7×10^{-20} W·Hz^{-1} in the latter case, we find that for $\Delta f = 100$ MHz, we have $Q_1 = 10^{-10}$ W and $Q_2 = 7\times10^{-12}$ W. Thus, a heterodyne detector is more sensitive. However, the useful range of a heterodyne detector is in practice very limited by the need to record CO_2 laser radiation and, therefore, it by no means excludes the use of parametric devices.

TABLE IX. Summary of typical experimental values of noise equivalent power (NEP) in parametric up-conversion of infrared signals (s is the sum frequency and d is the difference frequency).

λ_2, μ	λ_1, μ	λ_3, μ	Nonlinear crystal	η_e	NEP W·Hz$^{-1/2}$	References
1.7	0.6943	0.49 (s)	$LiNbO_3$	$3.5\cdot10^{-2}$	$5\cdot10^{-6}$	64
3.4	0.6328	0.53 (s)	$LiNbO_3$	$3.2\cdot10^{-6}$	$3\cdot10^{-8}$	66
10.6	0.6328	0.67 (d)	HgS	$6.3\cdot10^{-9}$	$9\cdot10^{-6}$	107
10.6	1.06	0.967 (s)	Ag_3SbS_3	$6.6\cdot10^{-8}$	$8\cdot10^{-7}$	149
3	0.5145	0.44 (s)	$LiNbO_3$	$8\cdot10^{-4}$	10^{-14}	69
3.4	0.6943	0.58 (s)	$LiJO_3$	5	$7\cdot10^{-16}$	91
10.6	0.647	0.69 (s)	Ag_3AsS_3	$1.5\cdot10^{-6}$	10^{-10}	35
2.9	0.488	0.42 (d)	$LiNbO_3$	$1.6\cdot10^{-3}$	$7.3\cdot10^{-13}$	62
1.15	1.064	0.55 (s)	$LiIO_3$	0.8	$1.5\cdot10^{-13}$	101

Applications of parametric detectors in optical communication systems with a carrier wavelength of 10μ are discussed in Refs. 144, 32, 3, and 13. The conclusion reached is that at present it is preferable to use direct detection for television purposes but parametric devices may be more convenient for transmission of telephone conversations and particularly of spectroscopic data.

We can thus identify the following promising fields of application of parametric infrared radiation detectors: 1) detection of weak infrared signals; 2) analysis of fast processes; 3) transfer of information at a carrier wavelength of 10μ, which does not require a very wide pass band.

4. PARAMETRIC CONVERSION OF INFRARED IMAGES

a) Brief review of main results

The present stage of research on parametric conversion of infrared images to the visible range started with the work of Midwinter.[65] He made visible, by means of an $LiNbO_3$ crystal ($\lambda_1 = 0.6943\mu$), an image of a linear grating formed at infinity in the radiation field of a xenon lamp ($\lambda_2 = 1.6\mu$). In an angular aperture of 60 mrad the number of resolved linear elements was $N_l \sim 50$. In 1968, Warner[112] was the first to achieve similar infrared conversion from the 10.6μ range in proustite ($\lambda_1 = 0.6943\mu$), which was characterized by an angular resolution of 15 mrad and by $N_l = 20$. Other investigations of basic importance were those carried out by Voronin *et al.* in 1969: they built the first parametric infrared holography system.[4,5] In particular, they made visible a three-dimensional hologram of a nearby object with a resolution close to the diffraction limit, which provided essentially a new effective method in infrared holography.

Studies of various laws governing parametric conversion of infrared images and ways of improving image characteristics started on a wide scale in 1969. Relative positions of infrared and converted images, longitudinal and transverse resolution in the best-focusing plane, influence of radiation divergence, and other associated questions were investigated theoretically and experimentally by a number of authors independently in the years 1969 to 1970. This work included a series of papers by Firester[55,56,67,68,190] and the work of Weller and Andrews,[46] Voronin, Il'inskiĭ *et al.*,[25,26] and others. Il'inskiĭ and Yanaĭt[26] introduced a scatter function, defining a field of a point infrared source, which has played a very fruitful role in the theoretical studies. Other calculation methods were based on ray paths;[11,46,100,190-192] their advantages are simplicity and clearness. This approach does not yield the spatial distribution of the image field but it does give the position and size of the image, etc. A summary of this first stage of investigations was given by Andrews.[11] It was soon found[25,26] that the Midwinter scheme with a distant object[21,65] is, in fact, less convenient than a scheme with a nearby object because the latter not only gives the same conversion efficiency and resolution, but also can be used to obtain three-dimensional images, is less critical to the divergence of the pump radiation and dis-

tortions of its wavefront, is less affected by inhomogeneities of the crystals, etc.

Theoretical and experimental investigations of the transverse resolution $\Delta_\perp$ outside the best-focusing plane and of the longitudinal resolution $\Delta_\parallel$ were reported in Refs. 51 and 71. In the case of a KDP crystal of thickness 6 mm ($\lambda_1 = \lambda_2 = 1.06\mu$) it was found that $\Delta_\perp = (2.5-2) \times 10^{-2}$ mm and $\Delta_\parallel = 11$ mm. Analytic expressions were obtained for the scatter functions and for $\Delta_{\perp,\parallel}$ in the near- and far-field Fresnel zones of an aperture stop. Subsequently, Kornienko, Ovechko, and Strizhevskiĭ[100] found approximate analytic expressions valid throughout the Fresnel zone.

Studies have been made of the influence of nonmonochromaticity.[102,111,193] In particular, it was shown in Ref. 111 that for a certain position of an infrared object, the role of chromatic aberrations is greatly reduced. Il'inskiĭ and Petnikova[193] showed that conversion of side components of nonmonochromatic radiation narrows the central peak of the scatter function and gives rise to wings. Investigations of the influence of the pump divergence have also been made (see also Refs. 102, 194, and 195). It was shown in Ref. 195 that in the case of critical phase matching and large values of $\Delta\omega_2$ the resolution is governed by the product of $\Delta\omega_2$ and the dispersion of the phase-matching directions but is independent of l. It follows from Refs. 196 and 102 that $\Delta_\perp$ hardly changes in going over to the range of high values of η_e and in the presence of absorption. However, according to Refs. 51 and 196, some increase in $\Delta_\perp$ can be achieved by a slight deviation from the direction of the tangential phase matching.

The transverse resolution is affected greatly by the inhomogeneity of a nonlinear crystal,[115,186,197] which results in angular broadening of the phase-matching curve and in corresponding deterioration of $\Delta_\perp$ by a factor of $(l/d)^{1/2}$ (d is the characteristic scale of the longitudinal inhomogeneity); the value of $\Delta_\perp$ does not change. Investigations of the image "speckle", which results from coherent illumination and reduces the resolution, is considered in Refs. 121 and 198. A method is suggested there for reducing this influence by small mechanical displacements of an object (or a scatterer, when a transparency is illuminated). In the pulse regime this effect results in averaging over the pulses.

In 1972 Chiou and Pace[118] constructed a system with an infinitely distant object pumped by the TEM_{00} mode alone ($\lambda_2 = 10.6\mu$, $\lambda_1 = 1.06\mu$, proustite) and achieved an angle of view 12° and an angular resolution 3 mrad, which was close to the diffraction limit (see also Ref. 132). Hulme and Warner[81] and Falk and Tiffany[117] developed a theory of parametric conversion of thermal infrared images and showed that, in particular, detection against a thermal background becomes more difficult as the number of resolved elements is increased. Lucy[27] carried out a series of careful measurements and demonstrated how the resolution deteriorates as the pump divergence is increased. Milton[12] (see also Ref. 13) analyzed thoroughly the functional applications of parametric conversion of infrared images and reached the conclusion that in the 10.6μ range the potentially most interesting applications are in viewing systems with laser illumination and in laser radar (lidar) with a relatively large number of resolved elements, but it should be remembered that careful development and optimization of the units is required because of the stiff competition offered by laser raster scanning systems.

The results reported above were obtained mainly using tangential phase matching. In 1970–1971, Gainer *et al.*[30,47,50] proposed and implemented a basically new scheme employing critical vector phase matching with strongly diverging pump radiation. The angular field of view is considerably wider in this scheme. Experimental implementation with a KDP crystal immediately gave an angular aperture of ~49° (Refs. 47 and 50). Theoretical and experimental investigations of critical vector phase matching were also reported elsewhere.[42,52,86,87,89,92,199-202] The method of classical Green's functions has been mainly used in the theory. It has been found[86,87,199] that if a cylindrical pump wave (the divergence is required only in the phase-matching plane) is used in critical vector phase matching, the resolution is governed by the diffraction of infrared radiation by the crystal aperture[5)] (in the case of a nearby infrared object there may be some aberrations, but they are easily suppressed by correcting optics with the one exception of distortion, which does not affect the resolution). A comprehensive comparison of the critical vector and tangential phase matching schemes can be found in Refs. 92 and 203.

The following image aberrations have been observed: astigmatism, which is the difference between the planes of best focusing along two mutually perpendicular directions due to the anisotropy,[50,96,100] geometric aberrations due to the complex shape of the phase-matching surface,[96,100,203] chromatic aberrations,[203] aberrations associated with lenses in the optical system,[204,205] and so on. Situations are possible in which aberrations not only lower the image quality but also limit the resolution.[96,97,200,201,203]

One should also mention some recent results. Tseng[123] demonstrated for the first time parametric conversion (in proustite) of an image of a distant object on a real (0.1–1 sec time scale using synchronized pulses from CO_2 and YAG:Nd^{3+} lasers. Harris and Bloom[158] showed that in the case of parametric conversion in metal vapors the width of the conversion band is fairly great for thermal images. Stappaerts, Harris, and Young[161] achieved conversion in cesium vapor of an infrared image (1000 elements) from the 2.9μ range to 4558 Å ($\eta_e = 20\%$). Molebnyĭ, Ovechko, and Strizhevskiĭ[105] reported intracavity conversion of an infrared image with an angular resolution of 1′–2′ (1.5–2 times greater than the diffraction limit) and a field of vision

[5)] If diffusely diverging pump radiation is used, the resolution is apparently governed by the angular width of the phase-matching curve (this has been initially assumed to apply in all cases[30,47]). However, in the tangential phase-matching case the resolution is governed by the thickness of the nonlinear crystal.

4–5° wide. Ustinov, Matveev, *et al.*[214-216] demonstrated the possibility of using nonlinear cyrstals in the formation of correlation, autocorrelation, and other image functions in real time.

b) Theory

The arrangement used in parametric infrared image conversion is shown schematically in Fig. 6. In a certain plane $z = z_2$, parallel to the faces of a nonlinear crystal NC, there is a given infrared field distribution ω_2. The ω_2 field emitted by this plane reaches the crystal, together with a pump wave ω_1, and is partly converted to the frequency $\omega_3 = \omega_1 + \omega_2$. We may assume formally that the field at the latter frequency is emitted from some plane $z = z_3$ on which the viewer's objective 0 is focused. The choice of this plane is to some extent arbitrary. The greatest interest lies in a plane ensuring the best transverse resolution of the image. By the resolution we mean the effective radius (along a given direction) of the image of a point infrared object.

There are various calculation methods for dealing with parametric conversion of infrared images. In the case of paraxial beams (for example, in the case of parametric conversion in a tangential phase matching scheme) the best effect is obtained by combining the method of truncated equations with the method of Fourier optics.[206] In the case of critical vector phase matching, when the divergence is very great, it is more effective to use the classical Green's functions.[86,87,200,201] Finally, we can also use the relatively simple and clear method of ray paths allowing for the interaction of waves in a nonlinear crystal, which has been used in the paraxial approximation.[11,46,100,190,191]

We shall now give some of the most important results of the theoretical calculations. The transverse and longitudinal resolutions $\Delta^{nc}_{\perp,\parallel}$ and the number of resolved (over an area) elements $N^{nc} = (N^{nc}_l)^2$ are calculated in Ref. 100 for the case of noncritical phase matching with a plane pump wave traveling along the z axis (Fig. 6) when an aperture stop with a Gaussian amplitude distribution of the transparency is placed in front of a crystal.

In particular, in the best-focusing plane and with an infrared object located in the "near-field" zone, we have

$$\Delta^{nc}_{\perp} \approx 1{,}4\sqrt{\frac{lk_1}{k_2k_3}}, \quad \Delta^{nc}_{\parallel} \approx l\frac{k_1}{2n_2k_3} \sim l,$$

and $N^{nc} \equiv (a_0/\Delta^{nc}_{\perp})^2 \sim a_0^2/\lambda_2 l$ in the same plane for an arbitrary value of z_2 (Refs. 25, 26, and 100); here, a_0 is the radius of the Gaussian stop or the radius of the pump beam. If $a_0 = l = 1$ cm and $\lambda_2 = 1\mu$, we have $N^{nc} \sim 10^4$. The longitudinal dimensions of an image increase by a factor of ω_3/ω_2 but the transverse dimensions are not affected. If the constant field approximation is not adopted, the value of $\Delta^{nc}_{\perp}$ deteriorates somewhat.[196,102] In the case of an infinitely distant infrared object it is useful to consider the angular field of view which in this case is proportional to ψ_2^{nc}. The angular resolution is of the order of the diffraction angle φ_{2d} and we have $N \propto (\psi_2^{nc} n_2/\varphi_{2d})^2$.

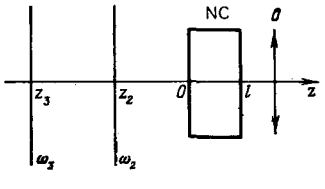

FIG. 6. Generalized diagram of a parametric infrared image converter.

In the case of critical vector phase matching we have $N^{cr} = (S_2/\lambda_2'^2)\varphi_1^2$, where S_2 is the area of the entry aperture of the converter, $\lambda_2' = \lambda_2/n_2$, and φ_1 is the pump divergence angle in the phase-matching plane.[87] For $S_2 = 1$ cm^2, $\lambda_2' = 1\mu$, and $\varphi_1 = 30°$, we have $N^{cr} = 2\times10^7$. This quantity is considerably greater than N^{nc} because of the rapid growth of the angular field of view, which is $\sim\varphi_1$ in any plane, whereas the angular resolution is still $\sim\varphi_{2d}$. The maximum value of φ_1 is limited by $2\theta_{pm} \sim 1$ rad $\gg \psi_2^{nc}$, where the index "pm" denotes phase matching. This restriction is governed by the condition of one-to-one correspondence between the beams at frequencies of ω_3 and ω_2 (Ref. 87). It should also be mentioned that realization of the maximum possible resolution in critical vector phase matching requires displacement of an infrared object to infinity, since in the opposite case there are geometric aberrations which lower the resolution.[86,98,100,203,207]

In the final analysis the principal characteristics of an image converter are the number of resolved elements N and the conversion efficiency η. A system ensuring the required value of N and also a high value of η is to be preferred. We shall, therefore, express η in terms of N for both systems. We shall find the dependence $\eta_e^{nc}(N) \propto N^{-2}$ by eliminating l from Eq. (4) and using $N^{nc} = (a_0/\Delta^{nc}_{\perp})^2$; the function $\eta_e^{cr}(N) \propto N^{-1/2}$ is obtained by eliminating φ_1 from the formulas $N^{cr} = (S_2/\lambda_2'^2)\varphi_1^2$ and $\eta_e^{cr} = BP_1/\varphi_1$ (the last formula is derived in Ref. 92, where B is a known constant). We shall assume that in both cases the entry pupil has the same area S. For moderately large values of N, η_e^{nc} decreases on increase of N much faster, we find that in the range $N \geqslant N_0$ we have $\eta_e^{cr} \geqslant \eta_e^{nc}$. Roughly speaking, we obtain $N_0 \propto 4(S/\lambda_2^2)^{2/3}$. Consequently, the critical vector phase matching system becomes more advantageous as we go deeper into the infrared range, i.e., for lower values of N. For example, if $S = 1$ cm^2, we find that $N_0 \approx 9\times10^5$ for $\lambda_2 = 1\mu$ and $N_0 \approx 4\times10^4$ for $\lambda_2 = 10\mu$.

The simplicity of the spatial wave decoupling in critical vector phase matching may make it useful[92,203] in the range $N \leqslant N_0$. It is also clear that for $N \leqslant 10^2$–10^3 (this is quite sufficient for many practical purposes), it is preferable to use tangential phase matching. The advantage of such matching is the retention of information on the volume of an infrared object.

c) Experimental aspects and applications

A typical experimental converter system based on tangential phase matching[111] differs from that shown in Fig. 1 only by an infrared transparency (which in the present case is a wire grid) in front of a lens 6. Its initial image at the wavelength of 10.6μ, formed by the germanium lens 6, may be located outside the crystal

FIG. 7. Wire-grid image made visible by conversion from 10.6 μ range:[109] a) width of infrared spectrum $\Delta_2 = 26$ cm^{-1}, $z_2 = 12$ cm (see Fig. 6); b) quasimonochromatic infrared radiation (CO_2 laser with one vibrational-rotational transition), $z_2 = 12$ cm; c) influence of nonmonochromaticity ($\Delta_2 = 26$ cm^{-1}) removed by suitable positioning of infrared object ($z_2 = l/2n_2$).

(on any side) or inside it. A plate 8 combining the beams $\omega_{1,2}$ is also made of germanium. The converted image is filtered and recorded with a photographic camera. Figure 7 shows photographs of the grid image for various positions of the infrared object and various widths of the infrared spectrum. In particular, we can see that the broadening of the infrared spectrum results in deterioration of the image quality (compare Figs. 7a and 7b) but this effect can be suppressed by a suitable selection of the object position (Fig. 7c).

The system based on critical vector phase matching is different. A typical system, taken from Ref. 87, is shown in Fig. 8. Interacting beams ($\lambda_{1,2} = 1.064\mu$) are directed to a nonlinear $LiIO_3$ crystal at the vector phase-matching angle (40°). A microslit and an objective forming a cylindrical image (in this case beyond the crystal) are placed in the path of each beam. The converted image is also located beyond the crystal and is recorded on a photographic film. The divergence plane is perpendicular to the optic axis; the working faces of the crystal are normal to the central rays. Figure 9 shows a converted image of a standard test pattern with an angular resolution of 28″ in the focusing plane and a linear resolution of 130 lines/mm.

An intracavity infrared image converter[105] is shown in Fig. 10. A nonlinear crystal 8 is placed inside the cavity (resonator) of a master laser oscillator ($\lambda_1 = 1.064\mu$, $oo \rightarrow e$) and this cavity is formed by mirrors 1 and 10; a Pockels cell is denoted by 2 and an active element by 3.

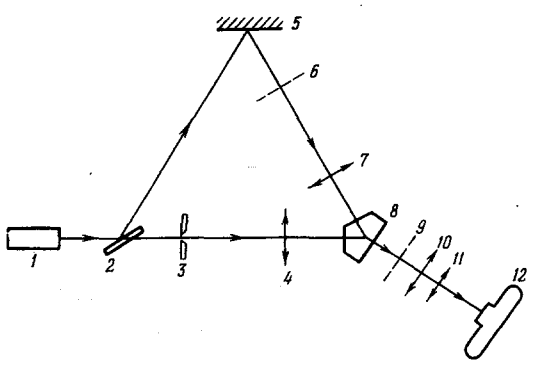

FIG. 8. Block diagram of a parametric infrared image converter utilizing critical vector phase matching:[87] 1) YAG:Nd^{3+} laser; 2) beam splitter; 3) microslit; 4), 7), 10), 11) objectives; 5) reflecting mirror; 6) infrared transparency; 8) $LiIO_3$ crystal; 9) position of best-focusing plane of visualized image; 12) photographic camera.

FIG. 9. Image of standard test pattern obtained by critical vector phase matching.[87]

Part of the radiation is deflected from the cavity by a plane-parallel plate 5 and is used as an infrared signal which is directed to a nonlinear crystal ($LiIO_3$, c axis perpendicular to the plane of the figure) by means of a rotatable mirror 6 and on the way it passes through a transparency $a-a'$; alternatively the mirror can be replaced by a diffusely reflecting object whose image is to be visualized; 11 is a filter and 12 is a photographic camera. The influence of losses in the nonlinear crystal plate 5 and elsewhere on the operation of the master laser oscillator is minimized by the use of intermediate feedback: an auxiliary mirror 4 is placed inside the cavity and the reflection coefficient of this mirror is optimized ($R_{opt} \sim 0.3$). The spatial structure of the field in the region of the crystal is controlled by placing the latter between two cylindrical lenses 7 and 9. Figure 11 shows a visualized image of a 1-ruble coin. The quality of the image is sufficient for practical purposes.

The main experimental results are summarized in Table X. We can see that systems with collinear and tangential phase matching can ensure typically an angular resolution of a few millirad. This resolution is an order of magnitude better in the case of critical vector phase matching. The vector method is characterized also by a larger angular field of view and a higher number of resolved elements. However, as pointed out above, it is inferior to the tangential phase-matching method in respect of the conversion efficiency, except in the range of high values of N.

Comparisons of parametric infrared image converters (for brevity, we shall call them parametricons) with other infrared viewers can be found in Refs. 10, 12, and 13. The devices already available are superior to parametricons in respect of thermal contrast detection. The resolution obtainable in collinear and tangential phase-matching systems is approximately the same (or slightly less) than that in the existing infrared viewers characterized by $N \sim 10^4 - 10^5$. However, in the case of critical vector phase matching it has been possible to achieve visualization with $N = 10^6$ (Refs. 30, 50, and 52)

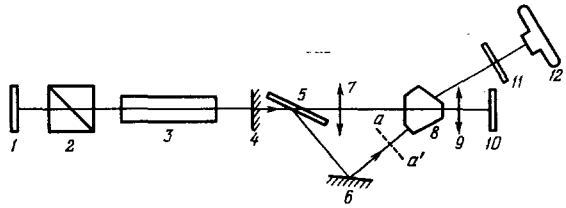

FIG. 10. Schematic diagram of an intracavity parametric image converter.[105]

FIG. 11. Image of a 1-ruble coin visualized inside a laser cavity.

and this is apparently not the maximum value which is possible: according to theoretical estimates,[87] it should be possible to achieve $N \approx 2 \times 10^7$. Therefore, parametricons based on critical vector phase matching are quite promising and competitive in those cases when very high values of N are required. Moreover, in the short-pulse regime, parametricons should be without competitors in any scheme.

5. NONLINEAR INFRARED SPECTROSCOPY

It is well known that spectrometers for the visible range have a number of important advantages over infrared spectrometers. Consequently, parametric conversion of infrared radiation to the visible range and subsequent spectral analysis of the converted radiation by methods which have reached a high stage of development in the visible range may lead to the development of efficient nonlinear infrared spectrometers.

Nonlinear infrared spectroscopy[6)] was first realized by Midwinter and Warner in 1967 (Ref. 64); they used $LiNbO_3$ ($\lambda_1 = 0.6943\mu$) to visualize infrared radiation from a mercury lamp in the 1.65–1.90μ range, which was covered by thermal tuning of phase matching in the range 19–200°C. In the visible range the line width was 17 Å (5–6 cm^{-1}). Spectral selectivity of ~10 cm^{-1} near $\lambda_2 \sim 1.6\mu$ was also reported in Ref. 65 and selectivity of ~1–5 cm^{-1} was found in the 1.6–3μ range in Ref. 61.

Thermal tuning is not always possible or effective. Khokhlov, Akhmanov, *et al.*[39,40] suggested a nonlinear infrared spectrometer with tuning by rotation of a crystal, which was subsequently realized by Midwinter.[31] Volosov[41] suggested the use of dispersion of the phase-matching angle for angular scanning of the second harmonic spectrum and he built a "panoramic" nonlinear spectrometer based on a KDP crystal and giving angular dispersion of 0.4 min/Å ($\lambda_1 = 1.06\mu$). Moreover, analysis of addition of unequal frequencies gave information on the time overlap of the various spectral components. Andreev, Volosov, and Kalintsev[77] showed that a nonlinear spectrometer based on $LiNbO_3$ ($\lambda_1 = \lambda_2 = 1.06\mu$) ensured linear dispersion of ~0.5 Å/mm (at 80°C), which was an order of magnitude better than that attainable in standard infrared prism spectrographs. The panoramic reception itself (on, for example, a photographic film), basically new in the infrared range,

TABLE X. Typical properties of image converters.

λ_2, μ	λ_1, μ	Method*)	Nonlinear crystal	Angular resolution, mrad	Angular field of view, **) deg	Linear resolution, lines/mm	Total number of resolved line elements	Total number of resolved area elements	References
1.6	0.6943	a	$LiNbO_3$	1.2	3.42		50		21, 65
1.15	1.064	a	KDP			28			190
10.6	0.6943	b	Ag_3AsS_3	15	17.1		20		20, 112
1.15	1.064	b	KDP	1.7 of diffr. limit					46
1.06	1.06	b	KDP			18—20	150	150×150	5, 25
1.06	0.6943	c	KDP	0.3—0.45	49	52	10^3	$10^3 \times 10^3$	50, 52
1.06	1.06	b	KDP		2	25			51
10.6	0.6943	b	Ag_3AsS_3		9.7	10			115
10.6	1.064	b	Ag_3AsS_3	3	12				118
10.6	1.064	b	$AgGaS_2$			13			141
10.6	1.064	c	Ag_3AsS_3	2.3			14	14×3	127
1.06	1.06	c	Ag_3AsS_3	0.14					123

*Here, a denotes collinear phase matching, b–tangential phase matching, and c–critical vector phase matching.
**In the phase-matching plane.

was of considerable importance. Kolpakov, Krivoshechkov, and Stroganov[42] used an $LiIO_3$ crystal to achieve cw conversion of a wide (0.11μ) spectrum of near infrared range by rotating a crystal automatically; they obtained dispersion of 0.17 min/Å and a resolution of 10 cm^{-1}. In the ~10μ range the angular dispersion achieved was ~0.1 min/Å (Ref. 109). Oraevskiĭ *et al.*[61,70,74,75] reached the conclusion that resolution of ~0.25 cm^{-1} should be possible in the near infrared range. A resolution of the order of 1 cm^{-1} was achieved in this range[62,69,91]; Refs. 69 and 73 give reports of the application of this method to astrophysics. Riccius and Siemsen[137] converted about 100 CO_2 laser lines from the 5.2–5.4μ range.

Nonlinear infrared spectroscopy has now been extended[27,108-110,128,146] to the ~10μ range. In particular, $\delta_2 \sim 1$ cm^{-1} was reported in Ref. 27 and a resolution of 0.9 cm^{-1} (in proustite) was obtained in Ref. 109. This resolution is largely limited by the pump divergence and inhomogeneities in a nonlinear crystal;[109,128] Arumov *et al.*[109] regard it as realistic to obtain a resolution of 0.1 cm^{-1} if these factors are eliminated (one might usefully employ here the method of preliminary shaping of infrared radiation[209] by a device with angular dispersion much greater than $d\theta/d\lambda$).

The simplest application of nonlinear infrared spectroscopy is in measurements of the integrated intensities of converted lines with some method of tuning. The resolution σ is then governed by the spectral width of the phase-matching curve for frequency addition δ_2, i.e., the infrared spectrum is averaged over the interval δ_2. Therefore, nonlinear spectroscopy of this type is called the integrating method.[28] We can see from Fig. 4 taken from Ref. 28 that δ_2 varies in the range 1–100 cm^{-1}. A study of the influence of pump nonmonochromaticity and angular divergence of waves on σ is also reported in Ref. 28.

More complex but more informative application of a spectrum analyzer in the visible range is to measure not the integrated intensity but the spectral composition of the converted (and, therefore, infrared) radiation; in terms of Ref. 28, this corresponds to "local" (without averaging over the interval ~δ_2) nonlinear spectroscopy.

6)The term nonlinear spectroscopy is used also for very-high-resolution spectroscopy within Doppler-broadened lines of gases[208] (and also in other contexts), but the use of this term here can hardly be misleading.

Naturally, much better resolution can then be achieved. For example, measurements[135] of the absolute wavelengths of the CO_2 laser lines in the $00^0 1-10^0 0$ and $00^0 2-02^0 0$ bands can be measured to within $\sim 2\times 10^{-8}$. In one special case a resolution of 10^{-5} cm^{-1} etc. has been achieved.[210] A resolution of $\sim 10^{-2}$ cm^{-1} realized in Ref. 93 may become a typical value; it is essentially limited by the pump line width.

In the case of local nonlinear spectroscopy it is desirable to have the conversion band as wide as possible. This makes useful the critical vector phase-matching method with strongly diverging pump radiation, which has been employed[88,89] to achieve conversion of the $2.7-3.7\mu$ range in a single pulse of a Q-switched ruby laser. It is also pointed out in Refs. 88 and 89 that the whole transparency range of lithium iodate and proustite may be converted employing pump focusing angles of about 40 and 60°, respectively. This method is very promising for recording wide infrared spectra, especially in the case of fast processes. It should be pointed out that considerable nonlinear distortions of the spectrum[93] are possible in the range $\eta_e \gtrsim 30\%$.

Table XI summarizes typical experimental results achieved in spectral resolution by nonlinear infrared spectroscopy. Clearly, the integrating nonlinear spectrometers are not inferior and local spectrometers are much superior in respect of resolution to the majority of conventional infrared spectrometers.

Another important advantage of nonlinear spectrometers is their fast response. The time constant of a parametric converter (i.e., the reciprocal effective width of the conversion band[109]) is $\sim 10^{-10}-10^{-12}$ sec, i.e., it is without any peers among the existing spectrometers. This is extremely important in fast-response infrared spectroscopy,[129,140,187] which at present is practicable under nanosecond (and especially picosecond) pulse conditions only by up-conversion of infrared radiation. The above discussion shows clearly the great promise of nonlinear infrared spectroscopy.

6. CONCLUSIONS

We can summarize the situation by stating that parametric up-conversion of infrared radiation is of considerable physical interest because many important relationships governing nonlinear optical processes are encountered in such conversion. Moreover, important practical applications can be expected in the infrared range, particularly in infrared signal detectors, infrared viewers, and nonlinear infrared spectrometers.

The results of investigations carried out so far provide convincing evidence of the promise and competitiveness of such devices, compared with existing instruments for the infrared range, but technical realization still requires further laboratory research.

Nonlinear infrared spectroscopy is especially promising because of its considerable advantages over conventional infrared spectroscopy (fast response and high spectral resolution are particularly marked). Specifically, the method of parametric conversion may become extremely important in picosecond infrared spectroscopy.

One of the central problems, on whose solution depends the general progress and further development of up-conversion, remains (as in other branches of quantum electronics and nonlinear optics) the search for new nonlinear media with high conversion efficiencies. It is desirable to combine work on improving the quality of the available crystals and search for new crystalline media with the mastering and practical utilization of gaseous nonlinear media. This essentially is still to be done. We cannot exclude the possibility that the use of gases and alkali metal vapors as nonlinear optical media[211] will result in a review of the earlier conclusions and appearance of new aspects of the problem under discussion. Intensive investigations are being carried out[212,213] but final conclusions would be premature.

The general progress in laser technology is also of considerable importance. The efficiency, reliability, simplicity of use, and availability of devices based on predetector parametric up-conversion of infrared radiation depend directly on the progress in the development and use of improved high-efficiency cw and pulsed lasers.

We would like to stress particularly the great personal contribution which was made to the subject by Academician R. V. Khokhlov. It suffices to mention work on infrared holography utilizing up-conversion of infrared radiation carried out with his personal participation, etc. His leadership and continuous encouragement have played a decisive role in the formulation and starting of a series of investigations of this subject at Moscow State University and other scientific centers in the Soviet Union. The present review was written on his initiative; he read the first version of the review and made many valuable comments. We acknowledge his help with gratitude.

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TABLE XI. Summary of typical spectral resolutions of nonlinear infrared spectrometers.

λ_2, μ	λ_1, μ	Type of spectrometer*	Nonlinear crystal	Spectral resolution, cm^{-1}	References
1.65—1.90	0.6943	i	$LiNbO_3$	5.4	64
10.6	0.6943	i	Ag_3AsS_3	5.4	110
1.6	0.6943	i	$LiNbO_3$	10	65
3	0.488	i	$LiNbO_3$	1	69
2.43—2.45	0.488	i	$LiNbO_3$	1.2	61
10.6	0.6943	i	Ag_3AsS_3	1	87
2.4—3.3	0.488	l	$LiNbO_3$	2.8	72, 76
3.39	0.6943	i	$LiIO_3$	2	80
1.06	1.06	l	$LiIO_3$	0.01	98
1.1	0.6328	i	$LiIO_3$	10	42
10.6	1.064	i	Ag_3AsS_3	1.8	108
2.4—3.4	0.488	i	$LiNbO_3$	2.9	62
10.6	0.6764	i	Ag_3AsS_3	11.7	126
1.6—3	0.6943	i	$LiNbO_3$	2.3	63
10.6	1.064	i,p	Ag_3AsS_3	0.9	109
10.6	1.064	l	GaSe	0.2	146

*Here, i, l, and p denote integrating, local, and panoramic nonlinear spectrometers, respectively.

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