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Phase matching of four-wave interactions of SRS components in birefringent SRS-active crystals

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Abstract. **A new method has been proposed for achieving wave vector matching in four-wave interactions of frequency components upon SRS in birefringent SRS-active crystals. The method ensures anti-Stokes wave generation and enables a substantial reduction in higher order Stokes SRS generation thresholds. Phase matching directions in $BaWO_4$ SRS-active negative uniaxial crystals and $SrWO_4$ SRS-active positive uniaxial crystals have been found in the wavelength range 0.4–0.7 μm.**

Keywords: *stimulated Raman scattering, four-wave mixing, wave vector matching, birefringent crystal.*

There is currently great interest in Raman lasers that provide a Stokes-shifted output. The use of high-performance crystalline SRS-active media is critical for the development of efficient and compact Raman converters and lasers. Among the most promising SRS-active materials are tungstates [$BaWO_4$, $SrWO_4$, $KGd(WO_4)_2$ and others], which offer high Raman gain and a broad transmission band (with a long-wavelength edge at ~5 μm) and can be doped with active laser ions [1, 2].

An important issue in the case of crystalline Raman lasers is the ability to ensure efficient generation of higher order SRS components and anti-Stokes waves. In the former case, a considerable increase in pump intensity (limited by the optical damage threshold of the crystal) is necessary, which would entail a drop in Raman gain coefficient with an increase in the wavelength of Stokes SRS components. In the latter case (anti-Stokes generation), the wave vector matching condition for four-wave coupling of SRS components should be satisfied, which is prevented by the high refractive index dispersion in crystalline media.

When the wave vector matching condition is satisfied, parametric four-wave mixing (FWM) ensures anti-Stokes wave generation through SRS and enables a substantial reduction in higher order Stokes SRS generation thresholds [3, 4]. In the nonlinear optics of quadratically nonlinear media [5], the wave vector matching condition for nonlinear interactions is exactly satisfied in a certain direction of a birefringent crystal upon interaction between orthogonally polarised waves. Here, we extend this method to nonlinear four-wave interactions of orthogonally polarised SRS components in birefringent SRS-active crystals with third-order nonlinearity.

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As shown previously [4], the contribution of parametric coupling to SRS depends on whether or not the wave vector matching condition is satisfied. In the case of partially degenerate FWM (PDFWM), we have for any three neighbouring SRS components

$$2\boldsymbol{k}_j = \boldsymbol{k}_{j-1} + \boldsymbol{k}_{j+1}. \quad (1)$$

In the case of nondegenerate FWM (NDFWM), a similar condition holds for any four neighbouring SRS components:

$$\boldsymbol{k}_j + \boldsymbol{k}_{j+1} = \boldsymbol{k}_{j-1} + \boldsymbol{k}_{j+2}. \quad (2)$$

Here, j is the number of the SRS component that is an FWM pump wave (an SRS pump wave at $j = 0$, anti-Stokes wave for $j < 0$ and Stokes wave for $j > 0$); $j - 1$, $j + 1$ and $j + 2$ are the numbers of the neighbouring SRS components involved in the FWM process under FWM pumping with the jth wave; and $\boldsymbol{k}_j$, $\boldsymbol{k}_{j-1}$, $\boldsymbol{k}_{j+1}$ and $\boldsymbol{k}_{j+2}$ are the wave vectors of these SRS components.

Refractive index dispersion in the medium leads to a wave vector mismatch in PDFWM,

$$\Delta k_j = k_{j-1} + k_{j+1} - 2k_j$$
$$= (n_{j-1} + n_{j+1} - 2n_j)2\pi\lambda_j^{-1} + (n_{j-1} - n_{j+1})2\pi\nu_R, \quad (3)$$

and a wave vector mismatch in NDFWM,

$$\Delta K_j = k_{j-1} + k_{j+2} - k_j - k_{j+1} = (n_{j-1} + n_{j+2}$$
$$- n_j - n_{j+1})2\pi\lambda_j^{-1} + (n_{j-1} + n_{j+1} - 2n_{j+2})2\pi\nu_R, \quad (4)$$

where n_j, n_{j-1}, n_{j+1} and n_{j+2} are the refractive indices for the respective waves; λ_j is the FWM pump wavelength; and ν_R is the Raman shift (expressed in inverse centimetres). This leads to violation of conditions (1) and (2) in the case of collinear generation of SRS components and reduces their parametric coupling.

Note that FWM pumping is provided by a wave at the initial frequency (wavelength λ_0) in FWM processes directed to the anti-Stokes region and by the first Stokes SRS component, which has a wavelength $\lambda_1 = (\lambda_0^{-1} - \nu_R)^{-1}$, in processes directed to the Stokes region.

Here, we seek conditions under which the wave vector mismatch (3) or (4) is zero, i.e., exact wave vector matching is ensured for FWM in SRS. Moreover, we are interested in how to ensure phase matching at any SRS pump wavelength and not only at the zero-dispersion wavelength [4]. For this reason, we propose realising FWM phase matching in a certain direction for orthogonally polarised waves, which is similar to the phase matching of three-wave interactions in the nonlinear optics of quadratically nonlinear media.

It is known [4, 6] that a steady-state model of collinear SRS with partially degenerate four-wave coupling between the SRS components can be represented by a system of equations for the SRS pump (E_0), anti-Stokes (E_{-1}) and Stokes (E_1) waves:

$$\frac{dE_{-1}}{dz} = -\frac{g}{2}\frac{\lambda_0}{\lambda_{-1}}|E_0|^2 E_{-1} - \frac{g}{2}\frac{\lambda_0}{\lambda_{-1}} E_0^2 E_1^* \exp(i\Delta k_0 z),$$

$$\frac{dE_0}{dz} = -\frac{g}{2}\frac{\lambda_1}{\lambda_0}|E_1|^2 E_0 + \frac{g}{2}|E_{-1}|^2 E_0, \qquad (5)$$

$$\frac{dE_1}{dz} = \frac{g}{2}|E_0|^2 E_1 + \frac{g}{2} E_0^2 E_{-1}^* \exp(i\Delta k_0 z),$$

where g is the Raman gain coefficient of the medium. The first terms in the second and third equations represent pump-to-Stokes SRS conversion, which is possible without FWM and does not require phase matching. For this reason, the Stokes wave usually has the same polarisation as the pump wave. The second terms in the first and last equations represent PDFWM coupling between the Stokes and anti-Stokes waves, which may lead to anti-Stokes wave generation, but only for $\Delta k_0 \to 0$. If the polarisation of the anti-Stokes wave is orthogonal to those of the Stokes and pump waves, we can achieve $\Delta k_0 = 0$ in some direction of a birefringent crystal and realise efficient collinear FWM anti-Stokes wave generation. Increasing the subscripts in (5) by unity, we obtain a system of equations for PDFWM in second Stokes wave generation. Just like above, when the polarisation of the second Stokes is orthogonal to those of the pump and first Stokes waves, we can achieve $\Delta k_1 = 0$ in a certain direction of the crystal and reduce the higher order Stokes generation thresholds. Similar reasoning applies to NDFWM.

Thus, we assume in what follows that the polarisation direction of the Stokes wave coincides with that of the SRS pump wave and that the wave resulting from FWM (anti-Stokes or higher Stokes component) is orthogonally polarised.

As birefringent SRS-active crystals we propose tungstates, typified by $BaWO_4$ (uniaxial negative) and $SrWO_4$ (uniaxial positive).

Let SRS-active crystals be oriented at angle Θ to their optic axis. In going from the light propagation direction along the optic axis (the direction corresponding to $\Theta = 0$) to an orthogonal direction ($\Theta = 90°$), the refractive index for extraordinary waves changes from n_o to n_e (n_o and n_e are the principal refractive indices of uniaxial crystals [5]). Θ can be found from the condition that the wave vector mismatch in the PDFWM process, Δk_j in (3), or that in the NDFWM process, ΔK_j in (4), be zero.

The refractive index for an extraordinary wave propagating at angle Θ is given by [5]

$$n^e(\Theta) = \frac{n_o n_e}{\sqrt{n_o^2 - (n_o^2 - n_e^2)\cos^2\Theta}}. \qquad (6)$$

The refractive index n^o for an ordinary wave propagating at angle Θ is independent of Θ and equals n_o.

Now, we can write expressions for the wave vector mismatch in FWM processes upon SRS when the waves resulting from the FWM are orthogonally polarised. From (3) and (6), we obtain expressions for the wave vector mismatch as a function of Θ in PDFWM upon SRS:

$$\Delta k_j^{ooe}(\Theta) = [n_{j-1}^e(\Theta) + n_{j+1}^o - 2n_j^o]2\pi\lambda_j^{-1} + [n_{j-1}^e(\Theta) - n_{j+1}^o]2\pi\nu_R,$$

$$\Delta k_j^{eeo}(\Theta) = [n_{j-1}^o + n_{j+1}^e(\Theta) - 2n_j^e(\Theta)]2\pi\lambda_j^{-1} + [n_{j-1}^o - n_{j+1}^e(\Theta)]2\pi\nu_R. \qquad (7)$$

From (4) and (6), we obtain expressions for the wave vector mismatch as a function of Θ in NDFWM upon SRS:

$$\Delta K_j^{oooe}(\Theta) = [n_{j-1}^e(\Theta) + n_{j+2}^o - n_j^o - n_{j+1}^o]2\pi\lambda_j^{-1} + [n_{j-1}^e(\Theta) + n_{j+1}^o - 2n_{j+2}^o]2\pi\nu_R,$$

$$\Delta K_j^{eeeo}(\Theta) = [n_{j-1}^o + n_{j+2}^e(\Theta) - n_j^e(\Theta) - n_{j+1}^e(\Theta)]2\pi\lambda_j^{-1} + [n_{j-1}^o + n_{j+1}^e(\Theta) - 2n_{j+2}^e(\Theta)]2\pi\nu_R. \qquad (8)$$

The Raman shift ν_R of $BaWO_4$ crystals is 926 cm^{-1} [7]. The Sellmeier equations for the dispersion curves of $BaWO_4$ crystals in the range $\lambda = 0.4-0.7$ μm have the form [8]

$$n_o^2 = 3.3550 + \frac{0.01738}{\lambda^2 - 0.08176} - 0.0965\lambda^2,$$

$$n_e^2 = 3.3460 + \frac{0.0177}{\lambda^2 - 0.08197} - 0.0962\lambda^2. \qquad (9)$$

Figure 1 shows the wave vector mismatches Δk_0^{ooe} and Δk_0^{eeo} calculated from (7) at $j = 0$ for anti-Stokes PDFWM and the mismatches ΔK_0^{oooe} and ΔK_0^{eeeo} calculated from (8) at $j = 0$ for anti-Stokes NDFWM as functions of SRS pump wavelength, λ_0, at the specified orientations, Θ, of a $BaWO_4$ crystal.

It can be seen from Fig. 1 that even at $\Theta = 90°$ there are SRS pump wavelengths at which the wave vector mismatch in FWM processes is zero. Exact phase matching occurs in ooe PDFWM at $\lambda_0 = 0.4371$ μm and in oooe NDFWM at $\lambda_0 = 0.4807$ μm. Note that the wavelength $\lambda_0 = 0.48$ μm (azur blue colour) can be provided by an argon pump laser. Anti-Stokes NDFWM upon SRS in a $BaWO_4$ crystal will then ensure anti-Stokes wave generation at $\lambda_{-1} = (\lambda_0^{-1} + \nu_R)^{-1} = 0.460$ μm (deep blue colour).

A decrease in Θ leads to an increase in the SRS pump wavelength at which the phase matching condition is satisfied for FWM upon SRS. A reduction in Θ to 53.23° ensures exact phase matching for anti-Stokes oooe NDFWM at the Nd:YAG second harmonic wavelength, $\lambda_0 = 0.532$ μm (Fig. 1b). At $\Theta = 36.35°$, there is exact phase matching for anti-Stokes ooe PDFWM (Fig. 1a) at the same wavelength $\lambda_0 = 0.532$ μm. Finally, at $\lambda_0 = 0.7$ μm (the long-wavelength boundary of the spectral range of our refractive index measurements) there is phase matching for anti-Stokes NDFWM and PDFWM processes at $\Theta = 30°$ (Fig. 1b) and 22.27° (Fig. 1a), respectively.

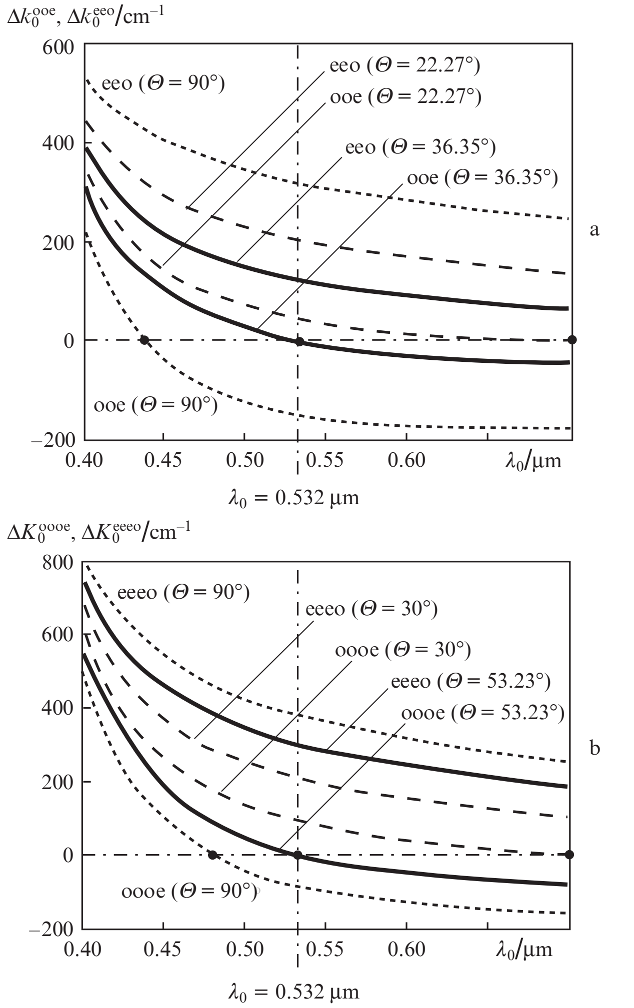

Figure 1. (a) Wave vector mismatches Δk_0^{ooe} and Δk_0^{eeo} for anti-Stokes PDFWM and (b) mismatches ΔK_0^{oooe} and ΔK_0^{eeeo} for anti-Stokes NDFWM as functions of SRS pump wavelength, λ_0, at the specified orientations, Θ, of a $BaWO_4$ crystal.

Note that the wave vector matching condition is satisfied in $BaWO_4$ crystals for an extraordinary anti-Stokes wave (generated through FWM). The other SRS components involved in this FWM process should be ordinary waves.

Similarly, the dispersion relations (9) can be used to obtain the wave vector mismatches Δk_1 and ΔK_1 for second Stokes PDFWM generation and third Stokes NDFWM generation, respectively. As a result, at $\Theta = 90°$ we have $\Delta k_1^{ooe} = 0$ (second Stokes PDFWM generation) and $\Delta K_1^{oooe} = 0$ (third Stokes NDFWM generation) at $\lambda_0 = 0.4138$ and 0.4539 μm, respectively.

To satisfy the wave vector matching condition for Stokes FWM upon SRS at a pump wavelength $\lambda_0 = 0.532$ μm, the angle Θ should be reduced to 45.27° for third Stokes NDFWM generation ($\Delta K_1^{oooe} = 0$) or to 33.59° for second Stokes PDFWM generation ($\Delta k_1^{ooe} = 0$). For the maximum wavelength $\lambda_0 = 0.7$ μm, we obtain phase matching for second Stokes PDFWM generation at $\Theta = 20.68°$ and that for third Stokes NDFWM generation at $\Theta = 19.44°$.

Thus, like in the case of anti-Stokes FWM, the phase matching condition for Stokes FWM is satisfied when the wave generated through FWM (second or third Stokes) is extraordinary and the other SRS components involved in this FWM process are ordinary waves.

The Raman shift ν_R of $SrWO_4$ crystals is 921.5 cm^{-1} [7]. The Sellmeier equations for the dispersion curves of $SrWO_4$ crystals in the range $\lambda_0 = 0.4–0.7$ μm have the form [8]

$$n_o^2 = 3.3219 + \frac{0.04538}{\lambda^2 - 0.007226} + 0.0257\lambda^2,$$

$$n_e^2 = 3.3490 + \frac{0.04825}{\lambda^2 - 0.0090808} - 0.03085\lambda^2. \quad (10)$$

Figure 2 shows the wave vector mismatches Δk_0^{ooe} and Δk_0^{eeo} calculated from (7) for anti-Stokes PDFWM and the mismatches ΔK_0^{oooe} and ΔK_0^{eeeo} calculated from (8) for anti-Stokes NDFWM as functions of SRS pump wavelength, λ_0, at the specified orientations, Θ, of a $SrWO_4$ crystal.

It is worth noting that the birefringence of $SrWO_4$ crystals [$n_e - n_o \approx 0.01$ according to (10)] is an order of magnitude higher than that of $BaWO_4$ crystals [$n_o - n_e \approx 0.002$ according to (9)]. As a result, no FWM phase matching can be achieved in $SrWO_4$ SRS-active crystals at $\Theta = 90°$. Only at a substantially smaller Θ, when the refractive index $n^e(\Theta)$ approaches

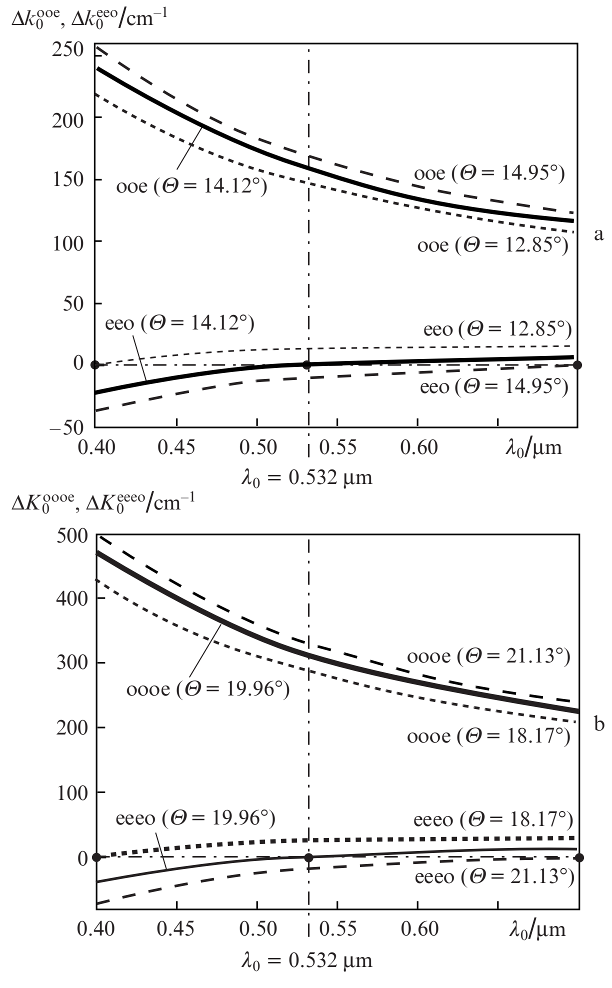

Figure 2. (a) Wave vector mismatches Δk_0^{ooe} and Δk_0^{eeo} for anti-Stokes PDFWM and (b) mismatches ΔK_0^{oooe} and ΔK_0^{eeeo} for anti-Stokes NDFWM as functions of SRS pump wavelength, λ_0, at the specified orientations, Θ, of a $SrWO_4$ crystal.

n_o, can the FWM phase matching condition be satisfied. Figure 2 shows the wave vector mismatches at $\Theta = 12.85°$ (Fig. 2a) and 18.17° (Fig. 2b), corresponding to the phase matching condition for anti-Stokes PDFWM and NDFWM processes, respectively, at an SRS pump wavelength $\lambda_0 = 0.4$ μm.

Notice that, in contrast to Fig. 1 (for $BaWO_4$ negative uniaxial crystals), where the ooe and oooe wave vector matching conditions are satisfied, Fig. 2 (for $SrWO_4$ positive uniaxial crystals) represents eeo and eeeo wave vector matching, which is the consequence of the inequality $n^e(\Theta) > n_o$. Just like above, a decrease in Θ leads to an increase in the SRS pump wavelength, λ_0, corresponding to FWM phase matching. At $\lambda_0 = 0.532$ μm, there is phase matching for anti-Stokes PDFWM generation at $\Theta = 14.12°$ (Fig. 2a) and that for anti-Stokes NDFWM generation at $\Theta = 19.96°$ (Fig. 2b). At $\lambda_0 = 0.7$ μm, the phase matching angles for the anti-Stokes PDFWM and NDFWM generation processes are $\Theta = 14.95°$ and 21.13°, respectively.

Similar calculations of the wave vector matching condition for Stokes FWM upon SRS in a $SrWO_4$ crystal using Eqns (9) and (10) at an SRS pump wavelength $\lambda_0 = 0.4$ μm give phase matching angles $\Theta = 14.07°$ and 19.85° for second Stokes PDFWM generation and third Stokes NDFWM generation, respectively. At $\lambda_0 = 0.532$ μm, we obtain phase matching at $\Theta = 15.46°$ and 21.98° for second Stokes PDFWM generation and third Stokes NDFWM generation, respectively. Finally, at $\lambda_0 = 0.7$ μm we have $\Theta = 16.44°$ and 23.63° for second Stokes PDFWM generation and third Stokes NDFWM generation, respectively.

It is worth pointing out that the phase matching condition for FWM upon SRS in $SrWO_4$ crystals, which have high birefringence, is satisfied in a broader wavelength range in comparison with $BaWO_4$ crystals, which have low birefringence. The orientation angle of $SrWO_4$ crystals varies relatively little, i.e. they have noncritical phase matching for FWM upon SRS. This allows one to use broadband laser radiation and ultrashort laser pulses for frequency conversion and to pump a particular SRS-active element by different lasers.

Thus, a new method has been proposed for achieving wave vector matching in four-wave interactions of frequency components upon SRS in birefringent SRS-active crystals. The method ensures anti-Stokes wave generation and enables a substantial reduction in higher order Stokes SRS generation thresholds.

Using the most promising SRS-active tungstate crystals as examples, we have shown that the wave vector matching condition for partially degenerate and nondegenerate four-wave mixing of SRS components is exactly satisfied in a certain direction of a birefringent crystal upon interaction between orthogonally polarised waves. The wave polarised orthogonally to the other SRS components is the anti-Stokes or a higher order Stokes component generated through FWM upon SRS.

Using known dispersion curves in the range 0.4–0.7 μm, we found the phase matching directions in $BaWO_4$ SRS-active negative uniaxial crystals and $SrWO_4$ SRS-active positive uniaxial crystals. Note that our calculations, performed in the plane wave approximation, are of interest for practical implementation of the effect in question in intracavity SRS, where no laser beam focusing is needed.

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