AZƏRBAYCAN MİLLİ ELMLƏR AKADEMİYASININ XƏBƏRLƏRİ
Fizika-riyaziyyat və texnika elmləri seriyası, 2003 №5(II)

# COHERENT POLARITON SCATTERING

**R.J.KASUMOVA, G.A.SAFAROVA**

*Baku State University*
*AZ 1148, Baku, Z.Khalilov str.23*

The theory of the coherent polariton scattering has been developed in the constant intensity approximation. In this approximation the inverse action of the excited wave on the pumping wave phase is taken into account. A comparison of the behavior of the polariton wave intensity for gallium phosphide obtained in the constant intensity approximation and the constant field approximation has been carried out.

Nonlinear spectroscopy methods are widely used for studying various excitations in gasses, liquids and solids. Spectroscopy of the coherent anti-Stokes scattering (CARS), and spectroscopy, based on registering of the Stokes waves amplification at SRS are the most used analysis methods. These methods are useful for medium excitation researches in noncentral symmetrical crystals, i.e., phonons in polar crystals [1].

Interest to CARS and SRS by polaritons is linked to their features, not conventional to the dissipation spectrums in non-polar excitations of the medium. Particularly, this is related to the property of polaritons to diffuse in the excitation medium to the macroscopic distances measuring from tens to hundreds of micrometers, especially, in enough distance from the dipole-active resonances, free path of polaritons can reach few centimetres. This property of polaritons was used for application of CARS spectroscopy at spatial diversion of the waves [2-5]. Those experimental researches are interesting from point of excitation transfer in the crystals [6].

As it is known, polariton is a material excitation of a mixed nature. Polaritons-based CARS and SRS spectroscopy is based on polaritons excitation in the field of strong exciting wave. Both processes can be researched using the coupled waves method, where in the most cases constant field approximation is used for the intensive exciting wave [7,8]. However, this approximation is valid only for the early stage of the interaction process, where the reverse impact of the excited polariton wave to the intense pump wave can be neglected. From experimental research on SRS it has been found that this process is accompanied with depletion of the excitation waves, which is inevitable at high levels of power or at long dimensions of the dissipating medium [7]. Thus, for researching waves interactions in non-linear medium it is recommended to use approximation of constant intensity [9], allowing consideration of reverse reaction of the excited waves on exciting waves. Another advantage of this approach is apart from the phase transformation of the interacting waves, losses of these waves can be registered as well. Latter is especially important for studying the processes with parametric character, which have threshold regime like coherent scattering.

This article reflects the results of further development of the theory of polariton scattering in the nonlinear medium with constant intensity approximation, and with consideration of the losses in interacting waves and presence of the phase mismatch between them.

Stimulated polariton scattering is described with the following equations system [8]

$$\frac{dA_p}{dz}+д_p A_p=-iг_{ps}^{(2)}A_s A_3\exp(-i\Delta z)-iг_{ps}^{(3)}\left|A_s\right|^2 A_p,$$
$$\frac{dA_s}{dz}+д_s A_s=-iг_s^{(2)}A_p A_3^*\exp(i\Delta z)-iг_s^{(3)}\left|A_p\right|^2 A_s, \qquad (1)$$
$$\frac{dA_3}{dz}+д_3 A_3=-iг_3 A_p A_s^*\exp(i\Delta z),$$

where $A_{p,s,3}$ are complex amplitudes of the exciting waves, Stokes component and polariton respectively, $щ_p, щ_s, щ_3$ are frequencies of the exciting wave, Stokes component and polariton wave, $д_j, г_j$ are absorption and nonlinear wave coupling indices of the waves.

$$г_{ps}^{(2)}=\frac{2\pi\omega_p^2}{k_{pz}c^2}\left(ч_{эфф}^{(2)}(щ_s)\right)^*,\quad г_{ps}^{(3)}=\frac{2\pi щ_p^2}{k_{pz}c^2}\left(ч_R^{(3)}(щ_s)\right)^*,\quad г_s^{(2)}=\frac{2рщ_s^2}{k_{sz}c^2}\left(ч_{эфф}^{(2)}(щ_s)\right),$$
$$г_s^{(3)}=\frac{2\pi\omega_s^2}{k_{sz}c^2}ч_R^{(3)}(щ_s),\quad г_3=\frac{2\pi щ_3^2}{k_{3z}c^2}ч_{эфф}^{(2)}(щ_3),\quad k_3=\frac{щ_з}{c}е_{3эфф}^{1/2}.$$

Here, $\chi_{эфф}^{(2)}$, $\chi_R^{(3)}$ are complex nonlinear susceptibilities, determined via matrix transition component, corresponding to the medium‘s excitation in the IR range and via parameters of the material excitation wave, $\chi_{эфф}^{(2)}$ is determined via conventional square non-linear susceptibility $\chi^{(2)}$. Condition for the frequencies of the interacting waves is following $щ_p=щ_s+щ_3$. Phase mismatch is determined from the expression $\Delta=k_{pz}-k_{sz}-k_{3z}$. Dispersion dependence of polariton is determined as $k_з=\frac{щ_з}{c}е_{3эфф}^{1/2}$ , where $е_{3эфф}^{1/2}=е_\infty+\frac{щ_o^2\Delta е}{щ_o^2-щ_3^2+i\Delta щ_L щ_o}$, $щ_o$ is transverse vibrations frequency of the crystal lattice. The constant term $е_\infty$ describes the response of the medium aside from the lattice vibrations, $\Delta е$, $\Delta щ_L$ are lattice parameters [10].

From (1) in approximation of the constant intensity, taking into account boundary conditions of $A_{p,s}(z=0)=A_{po,so}$, $A_3(z)=0$, for the intensity of the polariton wave it can be obtained

$$I_3^{SRS}(z)=\frac{г_3^2 I_{po}I_{so}}{x_1^2+y_1^2}\left(\sinh^2 x_1\cos^2 y_1+\cosh^2 x_1\sin^2 y_1\right)\exp\left[-(д_3+д_s)z\right], \qquad (2)$$

where

$$x_1=\sqrt{с_1}z\cos\frac{\varphi_1}{2},\quad y_1=\sqrt{с_1}z\sin\frac{\varphi_1}{2},\quad с_1^2=a_1^2+b_1^2,\quad \varphi_1=\mathrm{atan}(\frac{b_1}{a_1}),$$

$$a_1=\left(\frac{д_p+д_s+д_3}{2}\right)^2-\left(\frac{\Delta-г_{ps}^{(3)}I_{so}+г_s^{(3)}I_{po}}{2}\right)^2-г_3\left(г_{ps}^{(2)}I_{so}-г_s^{(2)}I_{po}\right),$$

$$b_1=\frac{д_p+д_s+д_3}{2}\left(г_{ps}^{(3)}I_{so}-г_s^{(3)}I_{po}-\Delta\right).$$

The process of the coherent scattering CARS can be described using the following system of equations [6,9]:

$$\begin{aligned}
&\frac{dA_p}{dz}+д_p A_p=-iг_{ps}^{(2)}A_s A_3\exp(-i\Delta z)-iг_{ps}^{(3)}\left|A_s\right|^2 A_p-iг_{pa}^{(3)}\left|A_a\right|^2 A_p,\\
&\frac{dA_s}{dz}+д_s A_s=-iг_s^{(2)}A_p A_3^*\exp(i\Delta z)-iг_s^{(3)}\left|A_p\right|^2 A_s,\\
&\frac{dA_a}{dz}+д_a A_a=-iг_a^{(2)}A_p A_3\exp\left[i(\Delta_a-\Delta)z\right]-iг_a^{(3)}A_p{}^2 A_s^*\exp\left(i\Delta_a z\right),\\
&\frac{dA_3}{dz}+д_3 A_3=-iг_3 A_p A_s^*\exp(i\Delta z),
\end{aligned} \tag{3}$$

where

$$г_a^{(2)}=\frac{2рщ_a^2}{k_{az}c^2}ч^{(2)}(щ_a),\ г_a^{(3)}=\frac{2\pi\omega_a^2}{k_{az}c^2}ч_R^{(3)}(щ_a),\ г_{pa}^{(3)}=\frac{2\piщ_p^2}{k_{pz}c^2}\left(ч_R^{(3)}(щ_a)\right)^*,\ \Delta_a=2k_p-k_s-k_a.$$

Solving (3) relatively to the polariton wave in the approximation of the constant intensity with consideration of the boundary conditions $A_{p,s,a}(z=0)=A_{po,so,ao},\quad A_3(z)=0,$ we get:

$$A_3^{CARS}(z)=-\frac{iг_s^{(3)}A_{po}A_{so}^*}{q_1^{CARS'}}\sinh q_1^{CARS'}z\exp\left(-\frac{p^{CARS'}}{2}z\right)\ \text{for}\ \left(p^{CARS'}\right)^2>4q^{CARS'}, \tag{4}$$

$$A_3^{CARS}(z)=-\frac{iг_s^{(3)}A_{po}A_{so}^*}{q_2^{CARS'}}\sin q_2^{CARS'}z\exp\left(-\frac{p^{CARS'}}{2}z\right)\ \text{for}\ \left(p^{CARS'}\right)^2\le 4q^{CARS'},$$

where

$$\left(q_1^{\mathrm{CARS}}\right)^2=\frac{\left(p^{\mathrm{CARS}'}\right)^2}{4}-q^{CARS'},\quad \left(q_2^{\mathrm{CARS}}\right)=q^{CARS'}-\frac{\left(p^{\mathrm{CARS}'}\right)^2}{4},$$

$$p^{\mathrm{CARS}'}=д_{\mathrm{p}}+д_3+\partial_s-i\left(\Delta-\left(г_{\mathrm{s}}^{(3)}\right)^*I_{po}-г_{\mathrm{ps}}^{(3)}I_{so}-г_{\mathrm{pa}}^{(3)}I_{ao}\right),$$

$$q^{CARS'}=г_3\left(г_{\mathrm{ps}}^{(2)}I_{so}-г_{\mathrm{s}}^{(2)}I_{po}\right)+д_3\left(д_{\mathrm{p}}+д_s\right)-iд_3\left(\Delta+\left(г_{\mathrm{s}}^{(3)}\right)^*I_{po}-г_{\mathrm{ps}}^{(3)}I_{so}-г_{\mathrm{pa}}^{(3)}I_{ao}\right).$$

From (3) at $г_{\mathrm{ps}}^{(2)}=г_{\mathrm{ps}}^{(3)}=г_{\mathrm{pa}}^{(3)}=0$ we get the result for the constant field approximation.

The results of the numerical analysis of the polariton wave behavior near the lattice resonance in case of the SRS in the cubical crystal of gallium phosphide GaP, belonging to the point group $\bar{4}3\mathrm{m}$ are illustrated on the Figure. The crystal parameters are as follows: $\varepsilon_\infty=8{,}457;\ \omega_0=366\,\mathrm{cm}^{-1},\ \Delta\omega_L=1{,}1\,\mathrm{cm}^{-1},\ \Delta\varepsilon=1{,}725$ [10-13]. The results of the approximations of the constant intensity and constant field are illustrated too. Comparison of the curves, corresponding to the various approximations shows that consideration of the phase changes of all interacting waves lead to reduction in efficiency of the conversion process (see curves 1 and 2).

This implies that the reduction of the conversion efficiency occurs not just due to incremental losses near the lattice resonance but also due to reverse impact of excited waves to the pumping wave.

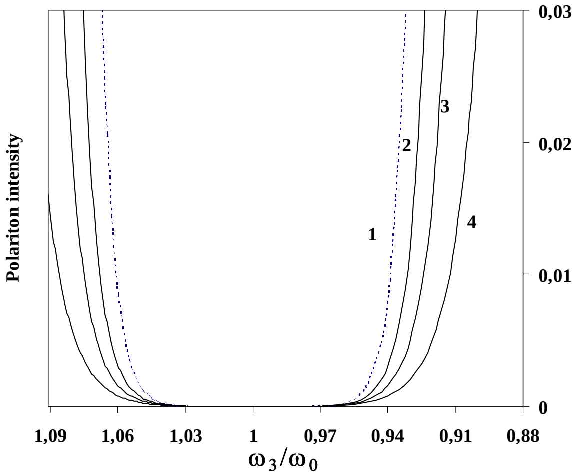


**Fig.**
Dependence of the polariton wave intensity $I_3 / I_{so}$ from polariton frequency $\omega_3 / \omega_0$ $\omega_0$=366cm$^{-1}$ in crystal GaP) for $\Delta$=0, $\delta_p$=$\delta_s$=0 at $I_{so}$ =0.3 (curves 1,2), 0.5 (curve 3) and 0.7 (curve 4), calculated in approximation of constant intensity (curves 2-4) and in approximation of the constant field (curve 1).


1. С.А. Ахманов, Н.И. Коротеев, *Методы нелинейной оптики в спектроскопии рассеяния света, Москва*, (1981).
2. F.Vallee, C.Flytzanis, *Phys. Rev. Lett.,* **74** (1995) 3281.
3. F.Vallee, G.M.Gale, C.Flytzanis, *Phys. Rev. Lett.,* **61** (1988) 2102.
4. F.Vallee, C.Flytzanis, *Phys. Rev. B*, **46** (1992) 1379.
5. T.Qiu, M.Maier, *Phys. Rev. B*, **56** (1997) 5717.
6. С.Н.Орлов, Ю.Н.Поливанов, *Квант. электрон.,* **31** (2001) 909.
7. Н.Бломберген., *Нелинейная оптика*, *Москва*, (1966) .
8. И.Р. Шен. *Принципы нелинейной оптики, Москва* (1989)
9. Z.H.Tagiev, R.J.Kasumova, R.A.Salmanova, N.V.Kerimova, *J. Opt. B: Quantum Semiclass. Opt.*, **3** (2001) 84.
10. Р.Пантел, Г.Путхоф, *Основы квантовой электроники*, *Москва*, (1972) 250.
11. C.H.Henry, C.G.B.Garrett, *Phys. Rev.,* **171** (1968) 1058.
12. W.L.Faust, C.H.Henry, *Phys. Rev. Lett.,* **25** (1966) 1265.
13. C.H.Henry, J.J.Hopfield, *Phys. Rev. Lett.,* **22** (1965) 964.


**KOHERENT POLARİTON SƏPİLMƏSİ**

**R.C.QASIMOVA, G.A.SƏFƏROVA**

**Koherent polariton səpilməsi nəzəriyyəsi sabit intensivlik yaxınlaşmasında inkişaf etdirilmişdir.Bu yaxınlaşmada həyəjanlaçmiş dalğanın güclü dalğanınfazasına əks təsiri nəzərə alınmışdir. GaP kristali üçün polaritondalğası intensivliyi hır iki sabit amplitud və sabit intensivlik yaxınlaşmalarında mügayisə edilmişdir.**

**КОГЕРЕНТНОЕ ПОЛЯРИТОННОЕ РАССЕЯНИЕ**

**Р.Дж.КАСУМОВА, Г.А.САФАРОВА**

Рассматривается теория когерентного поляритонного рассеяния в приближении заданной интенсивности. В этом приближении принимается во внимание обратная реакция возбуждаемой волны на волну накачки. Проведено сравнение поведений интенсивности поляритонной волны для кристалла GaP, полученные в приближениях заданной интенсивности и заданного поля.

Редактор: Г.Аждаров