

## TRANSFORM RELATIONSHIPS BETWEEN ABSORPTION AND COHERENT RESONANCE RAMAN SCATTERING IN HOMOGENEOUS AND INHOMOGENEOUS MEDIA

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**Abstract.** The amplitudes/excitation profiles of coherent resonance Raman scattering (CARS and CSRS) are directly related to the one-photon absorption. The effect of the inhomogeneity of solvent on the molecular spectra and the transform relationships is examined. The study is based on the transform theory applied in investigations of spontaneous resonance Raman scattering to relate the Raman excitation profile to the molecular absorption observed.

**Key words:** coherent resonance Raman scattering (CARS & CSRS), Raman excitation profiles, transform method, multimode systems, homogeneous and inhomogeneous media.

### INTRODUCTION

It is well known nowadays that the excitation spectrum (REP) of spontaneous resonance Raman scattering (RRS) can be represented via the transform of the (measured) absorption spectrum [1] (see also reviews [2, 3]). This approach — the calculation of REPs by the transform method — is a practical solution of the multimode problem in the RRS theory. The problem arises because all the Franck-Condon active modes take part in the RRS process. Therefore in a multimode case, like complex organic molecules or impurity centres in crystals, one should take into account a large number of parameters to get information about the coupling with the Raman-active mode. In the transform method [4–7], the multimode information is automatically carried through the calculation via the use of the measured absorption: the excitation profile of the Raman mode is computed directly from the measured absorption spectrum, while model parameters are needed only for the Raman mode.

These considerations also apply to coherent resonance Raman scattering like CARS (coherent anti-Stokes Raman scattering) and CSRS (coherent Stokes Raman scattering). In these processes, four photons are simultaneously involved: two photons of the «pump» frequency,  $E_p$ , are absorbed, and two photons of the Stokes and the anti-Stokes shifted frequency,  $E_p - \delta$  and  $E_p + \delta$ , are emitted. In a CARS experiment, the sample is subjected to laser beams of frequencies  $E_p$  and  $E_p - \delta$  while

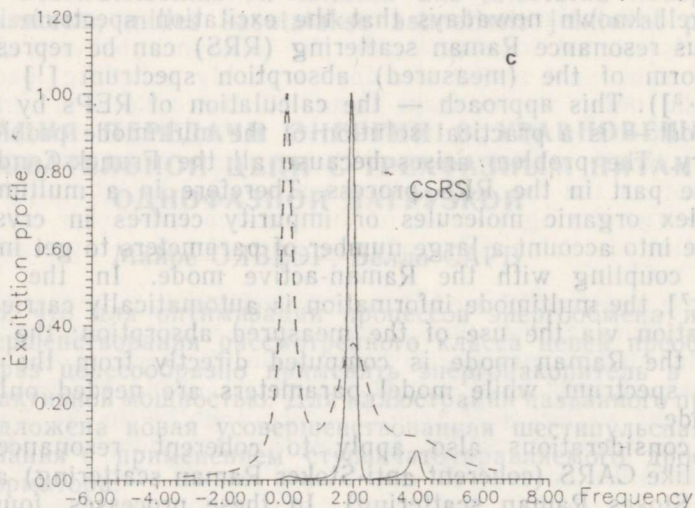
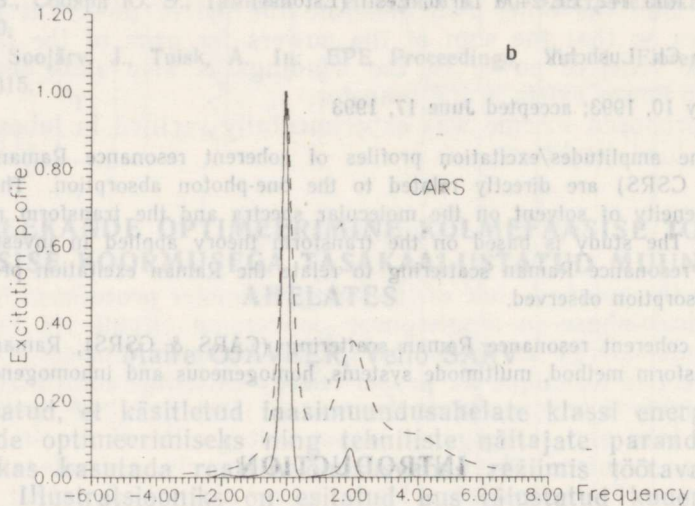
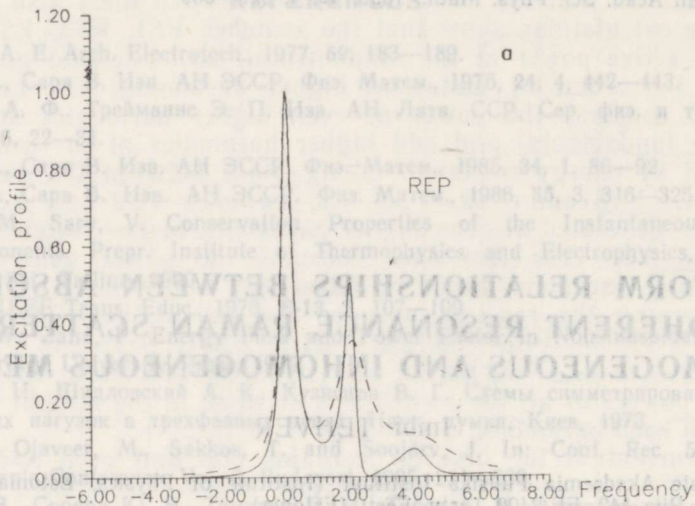


Fig. 1. Absorption spectrum according to Eq. (9) (dashed line) and Raman excitation profiles (solid lines),  $\gamma=0.2$ ,  $\Gamma=0.6$ ,  $\xi^2=1$ ,  $\omega=2$  (parameters are given in dimensionless units).

the signal beam at the anti-Stokes frequency  $E_p + \delta$  is monitored; a CSRS experiment differs in that the anti-Stokes frequency is incident upon the sample, and the Stokes  $E_p - \delta$  beam is monitored as the signal. In an excitation profile experiment, incident frequencies are scanned together so that their difference,  $\delta$ , is constant and equal to a Raman mode of the sample.

At  $T=0$  the absorption spectrum and the incoherent resonance Raman amplitude can be described by, respectively,

$$I_{\text{abs}}(\omega_0) = \text{Im} \sum_{m=0}^{\infty} \frac{\langle 0 | M_1 | m \rangle \langle m | M_1 | 0 \rangle}{\omega_0 - E_m - i\Gamma}, \quad (1)$$

$$\alpha(\omega_0) = \sum_{m=0}^{\infty} \frac{\langle 0 | M_1 | m \rangle \langle m | M_2 | 1 \rangle}{\omega_0 - E_m - i\Gamma}, \quad (2)$$

where  $|0\rangle$  and  $|1\rangle$  are the initial and final vibrational states in the ground electronic state,  $|m\rangle$  is the intermediate vibrational state in the excited electronic state and  $\Gamma$ , its decay constant;  $E_m$  is the energy of the state  $|m\rangle$ ,  $M_1$  and  $M_2$  are the electronic transition matrix elements for excitation and emission, and  $\omega_0$  denotes the excitation frequency.

For the CARS process we have [8,9]

$$\chi_{\text{CARS}} = \sum_m \frac{\langle 0 | M_1 | m \rangle \langle m | M_2 | 1 \rangle}{\omega_0 - E_m - i\Gamma} \sum_{m'} \frac{\langle 0 | M_3 | m' \rangle \langle m' | M_4 | 1 \rangle}{\omega_0 + \omega_j - E_{m'} - i\Gamma} \quad (3)$$

( $\omega_j$  is the Raman frequency). For CSRS,

$$\chi_{\text{CSRS}} = \sum_m \frac{\langle 0 | M_1 | m \rangle \langle m | M_2 | 1 \rangle}{\omega_0 - E_m - i\Gamma} \sum_{m'} \frac{\langle 0 | M_3 | m' \rangle \langle m' | M_4 | 1 \rangle}{\omega_0 - \omega_j - E_{m'} + i\Gamma}. \quad (3')$$

The excitation profiles are defined by the square of modulus of the third-order susceptibility  $|\chi|^2$ . Comparing Eqs. (2) and (3), one can see that

$$\chi_{\text{CARS}} = \alpha(\omega_0) \alpha(\omega_0 + \omega_j), \quad (4)$$

$$\chi_{\text{CSRS}} = \alpha(\omega_0) \alpha^*(\omega_0 - \omega_j). \quad (4')$$

It is easy to see (make the transform  $\omega_0 \Rightarrow \bar{\omega}_0 = \omega_0 + \omega_j$ ) that the excitation profiles of CARS and CSRS have the same shape, CSRS is just shifted to high-frequency side of CARS by the Raman mode  $\omega_j$  (Figs. 1b, 1c).

Spectral lines can be broadened by different kinds of mechanisms. A homogeneous mechanism (lifetime broadening) broadens the spectral lines of all the molecules equally, an inhomogeneous mechanism (solvent effect) spreads the molecular energy values over some mean value. Let us first look for the transform laws in homogeneous media.

### TRANSFORM RELATIONSHIPS BETWEEN ABSORPTION AND SCATTERING FOR IDENTICAL MOLECULES

The transform law between the absorption and excitation profiles of Raman scattering depends on the model of vibronic coupling. In systems obeying standard assumptions, like the adiabatic and Condon approximation, harmonic vibrations and linear vibronic coupling, the incoherent Raman amplitude is related to the absorption spectrum as follows [4,5]:

$$a_j(\omega_0) \sim \xi_j [\Phi(\omega_0) - \Phi(\Omega)], \quad (5)$$

where  $\Omega = \omega_0 - \omega_j$  is the frequency of the scattered light and  $\xi_j$  is the parameter of linear coupling ( $\xi_j^2$  are Stokes losses) of the Raman mode  $j$ .  $\Phi(\omega)$  is the complex refractive index determined by the optical absorption  $I_{\text{abs}}(\omega_0)$  as follows:

$$\Phi(\omega) = i\pi I_{\text{abs}}(\omega_0) + P \int_{-\infty}^{\infty} dx \frac{I_{\text{abs}}(x)}{x - \omega}. \quad (6)$$

The REP of the first-order spontaneous Raman scattering (Stokes component) is determined as

$$I_j^s(\omega_0) \sim \xi_j^2 |\Phi(\omega_0) - \Phi(\omega_0 - \omega_j)|^2. \quad (7)$$

The REP of the anti-Stokes incoherent RRS equals

$$I_j^a(\omega_0) \sim \xi_j^2 |\Phi(\omega_0) - \Phi(\omega_0 + \omega_j)|^2. \quad (7')$$

Eqs. (7) are the main relationships of the transform method. The first term of Eq. (7) describes the resonance with the excitation frequency, the second one corresponds to the resonance with the frequency of the scattered light, both together describe the interference of these two channels.

Taking into account Eqs. (4) and (7) we obtain directly the transform laws for CARS [10]:

$$I_{\text{CARS}} = |\chi_{\text{CARS}}|^2 \sim |[\Phi(\omega_0) - \Phi(\omega_0 - \omega_j)][\Phi(\omega_0 + \omega_j) - \Phi(\omega_0)]|^2, \quad (8)$$

and CSRS:

$$I_{\text{CSRS}} = |\chi_{\text{CSRS}}|^2 \sim |[\Phi(\omega_0) - \Phi(\omega_0 - \omega_j)][\Phi^*(\omega_0 - \omega_j) - \Phi^*(\omega_0 - 2\omega_j)]|^2. \quad (8')$$

Eqs. (7), (8) and (8') are used for model calculations of excitation profiles of spontaneous Raman scattering (REPs), CARS and CSRS when the absorption is chosen as

$$I_{\text{abs}}(\omega_0) = \frac{\gamma}{\omega_0^2 + \gamma^2} + \frac{\Gamma}{(\omega_0 - \bar{\omega})^2 + \Gamma^2} + \frac{\xi^4}{2!} \frac{2\Gamma}{(\omega_0 - 2\bar{\omega})^2 + 4\Gamma^2} \quad (9)$$

Figs. 1a—c demonstrate the results.

## TRANSFORM LAWS IN THE INHOMOGENEOUS CASE

In the coherent case the radiation scattered from different molecules interferes. In the spontaneous Raman scattering there is no such interference. Hence, the sequence of operations in performing the inhomogeneous broadening convolution is different for the coherent and the spontaneous Raman scattering. To include inhomogeneous broadening in the description of the incoherent spectrum, one must evaluate the integral

$$I = \int |a|^2 \rho(\epsilon) d\epsilon, \quad (10a)$$

where  $\rho(\epsilon)$  is the probability distribution function of molecular energy values. For coherent Raman processes like CARS and CSRS, the inhomogeneous convolution is performed before taking the absolute square [9]:

$$I_{\text{CARS}} = \left| \int \chi_{\text{CARS}} \rho(\epsilon) d\epsilon \right|^2. \quad (10b)$$

Further it is assumed that the inhomogeneities affect the energy of the electronic transition  $\Omega_{21}$  in the molecule but do not change its local dynamics. The model holds for systems where the inhomogeneous dispersion of the vibrational frequency is less than the homogeneous width of the vibrational level.

Let us use the Fourier amplitude approach as very convenient for a general solution of the problem. In this approach the absorption spectrum is given by the integral [11]

$$I_{\text{abs}}(\omega_0) = \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \Omega_{21})t} F(t), \quad (11)$$

where  $F(t)$  is the Fourier transform of the absorption.

Analogously we can represent the Raman amplitude [5]:

$$a(\omega_0) = \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \Omega_{21})t} A(t). \quad (12)$$

In the case of the basic model, the Raman Fourier transform is related to the absorption Fourier transform as follows [5]:

$$A(t) = i\Theta(t)\xi_j(1 - e^{-it\omega_j})F(t), \quad (13)$$

where  $\Theta(t)$  denotes the Heaviside step function which takes into account the causality law in RRS, i. e. scattering follows excitation.

Taking into account Eqs. (4) we obtain for CARS

$$\chi_{\text{CARS}} = \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \Omega_{21})t} A(t) \int_{-\infty}^{\infty} dt' e^{i(\omega_0 - \Omega_{21} + \omega_j)t'} A(t') \quad (14)$$

and CSRS

$$\chi_{\text{CSRS}} = \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \Omega_{21})t} A(t) \int_{-\infty}^{\infty} dt' e^{-i(\omega_0 - \Omega_{21} - \omega_j)t'} A^*(t'), \quad (15)$$

where in the case of the basic model the Fourier amplitude  $A(t)$  is defined via the absorption Fourier transform  $F(t)$  by Eq. (13).

Using Fourier transforms for the spectra and the distribution function  $\varrho(\epsilon) \equiv \varrho(\Omega_{21} - \bar{\Omega}_{21})$  ( $\bar{\Omega}_{21}$  denotes the mean pure-electronic transition frequency), we get, after the integration of (11) for the inhomogeneous absorption spectrum,

$$\begin{aligned} \mathcal{I}_{\text{abs}}(\omega_0) &= \int_{-\infty}^{\infty} d\Omega_{21} \varrho(\Omega_{21}) \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \Omega_{21})t} F(t) = \\ &= (2\pi)^{-1} \int_{-\infty}^{\infty} d\Omega_{21} \int_{-\infty}^{\infty} dt' e^{i(\Omega_{21} - \bar{\Omega}_{21})t'} \varrho(t') \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \Omega_{21})t} F(t) = \\ &= \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \bar{\Omega}_{21})t} F(t) \varrho(t) \end{aligned} \quad (16)$$

and for the inhomogeneous incoherent REP,

$$\begin{aligned} \mathcal{I}_R(\omega_0) &= \xi_j^2 \int_0^{\infty} \int_0^{\infty} dt dt' e^{i(\omega_0 - \Omega_{21})(t-t')} \times \\ &\times F(t) F^*(t') (1 - e^{-i\omega_j t}) (1 - e^{-i\omega_j t'}) \varrho(t - t'). \end{aligned} \quad (17)$$

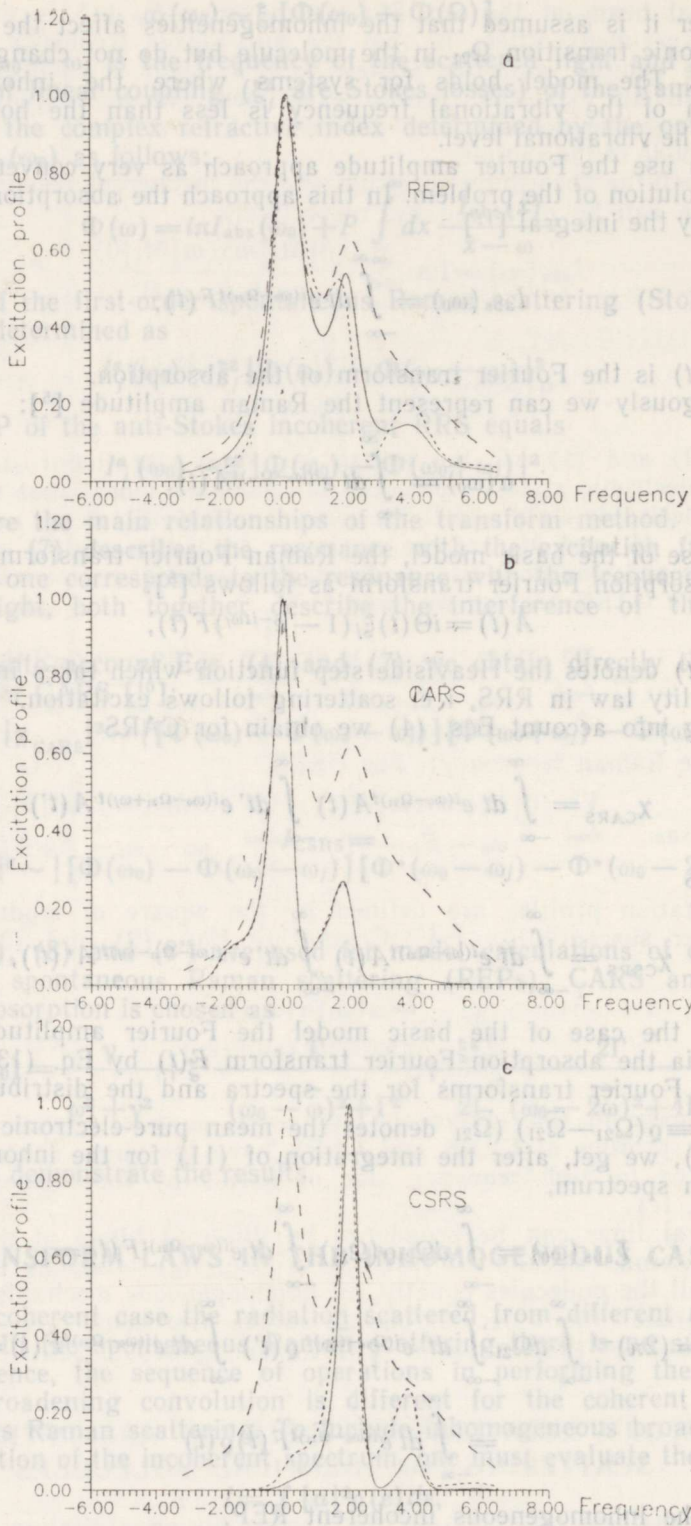


Fig. 2. Absorption spectrum (dashed line) and Raman excitation profiles calculated by Eqs. (7) and (8) (dotted lines) and by Eqs. (17) and (18) (solid lines, exact result) with taking into account the inhomogeneous broadening of the Lorentzian distribution

$$(\Delta_L=0.33), \gamma=0.2, \Gamma=0.6, \xi^2=1, \bar{\omega}=2,$$

Here  $\omega_j$  is the Raman frequency and  $\rho(t)$  denotes the Fourier transform of the distribution of pure-electronic transition frequencies. In the case of a Gaussian distribution  $\rho(t) = \exp(-t^2\Delta_G^2/2)$ , for a Lorentzian distribution  $\rho(t) = \exp(-\Delta_L|t|)$ . When comparing Eqs. (16) and (17) one can see that (17) cannot be factorized to the basic transform law (7) as it was in the homogeneous case because of the mixing to two integrals by  $\rho(t-t')$ . It is shown in Fig. 2a (Lorentzian distribution) and Fig. 3a (Gaussian distribution): in the case of inhomogeneously broadened absorption (dashed curve) the REP calculated according to the transform law (7) (dotted curve) does not follow the shape of the correct REP (Eq. (17), solid line).

Taking into account Eqs. (10b) and (14), we find in an analogous way for CARS:

$$I_{\text{CARS}} = \left| \xi_j^2 \int_0^\infty \int_0^\infty dt dt' e^{i(\omega_0 - \bar{\Omega}_{21})(t+t')} F(t) F(t') \times \right. \\ \left. \times (1 - e^{-i\omega_j t}) (e^{i\omega_j t} - 1) \rho(t+t') \right|^2 \quad (18)$$

and CSRS:

$$I_{\text{CSRS}} = \left| \xi_j^2 \int_0^\infty \int_0^\infty dt dt' e^{i(\omega_0 - \bar{\Omega}_{21})(t-t')} F(t) F^*(t') \times \right. \\ \left. \times (1 - e^{-i\omega_j t}) (e^{i\omega_j t} - e^{2i\omega_j t'}) \rho(t-t') \right|^2. \quad (18')$$

Looking at Eqs. (18), some general conclusions can be drawn immediately. For a Lorentzian inhomogeneous distribution in CARS

$$\rho(t+t') = \exp[-\Delta_L(t+t')] = \rho(t)\rho(t'), \quad (t, t' > 0)$$

consequently Eq. (18) can be factorized due to what the basic transform law (8) holds in the inhomogeneous case as well (Fig. 2b: CARS calculated by Eqs. (8) and (18) coincide). The linewidths in the CARS spectrum depend, analogously to absorption, on the sum of homogeneous and inhomogeneous widths. An analogous result was obtained earlier by Desiderio and Hudson [8] but only for the case of the absorption spectrum of the Lorentzian shape. Here it is shown for an arbitrary absorption spectrum. However, such a factorization is not quite correct for a Gaussian inhomogeneous distribution (Fig. 3b: note that there is a much smaller difference between the two curves than in the case of REP or CSRS).

In contrast to CARS, CSRS Eq. (18') can be factorized neither in the case of a Lorentzian distribution (Fig. 2c) nor for a Gaussian distribution (Fig. 3c), similarly to the spontaneous Raman scattering (compare with Eq. (17)). Transform law (8') does not hold for the inhomogeneous case. Note also that in the case of inhomogeneous broadening the excitation profile of CSRS is sharper than that of CARS. In contrast to the homogeneous case, the excitation profiles of CARS and CSRS no longer have the same shape.

In the case of the Gaussian distribution of pure-electronic frequencies, when the inhomogeneous absorption spectrum is determined by

$$I_{\text{abs}}(\omega_0) = \int_{-\infty}^{\infty} dt e^{i(\omega_0 - \bar{\Omega}_{21})t} F(t) \exp(-t^2\Delta_G^2/2), \quad (19)$$

the inhomogeneous REP,

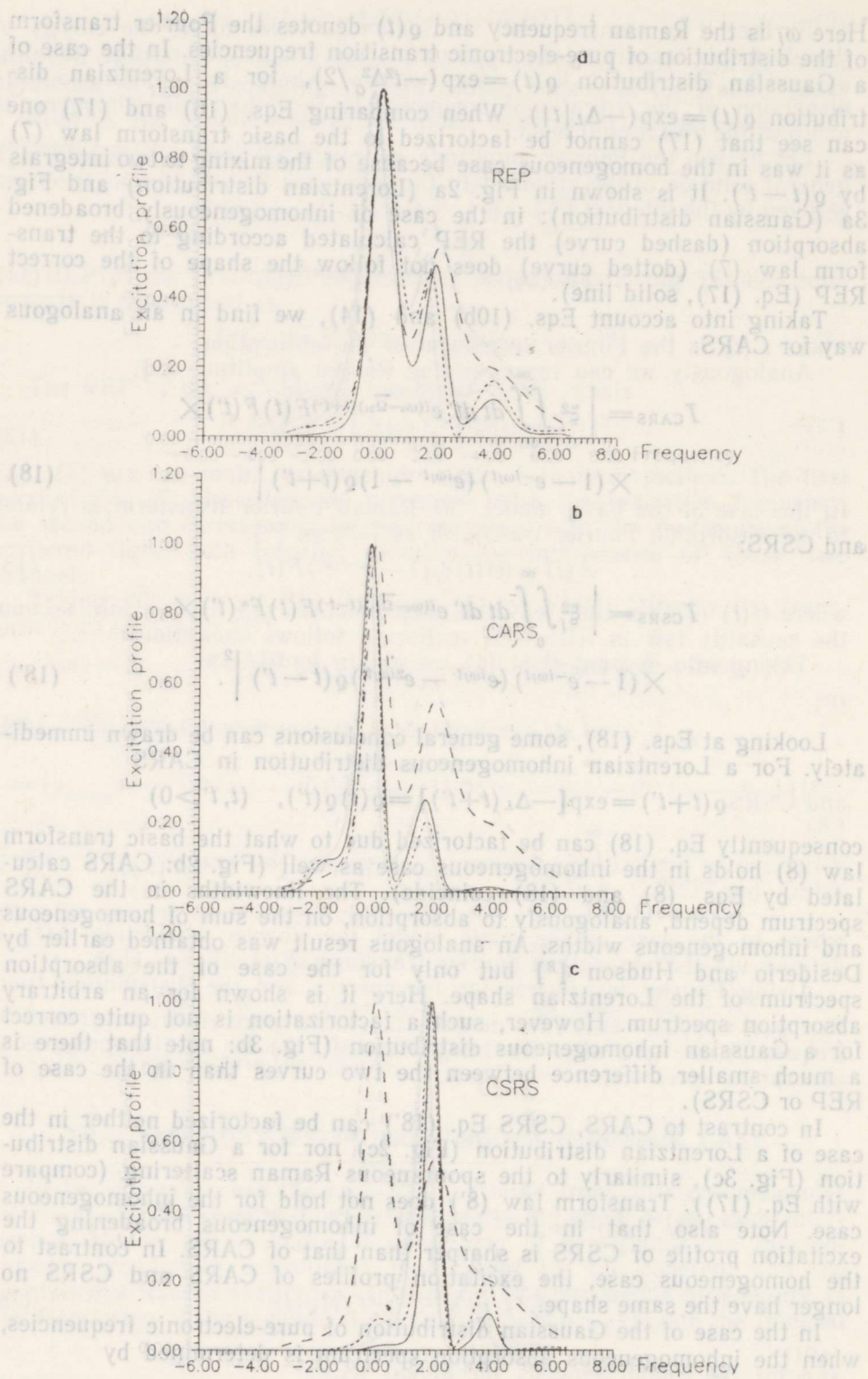


Fig. 3. Absorption spectrum (dashed line) and Raman excitation profiles calculated by Eqs. (7) and (8) (dotted lines) and by Eqs. (17) and (18) (solid lines, exact result) with taking into account the inhomogeneous broadening of the Gaussian distribution ( $\Delta\omega=0.28$ ).  $\gamma=0.2$ ,  $\Gamma=0.6$ ,  $\xi^2=1$ ,  $\bar{\omega}=2$ .



$$\begin{aligned} \tilde{I}_R(\omega_0) = & \xi_j^2 \int_0^\infty \int_0^\infty dt dt' e^{i(\omega_0 - \bar{\Omega}_{21})(t-t')} F(t) F(t') \times \\ & \times (1 - e^{-i\omega_j t}) (1 - e^{i\omega_j t'}) e^{-(t-t')^2 \Delta_G^2 / 2}, \end{aligned} \quad (20)$$

transforms after the expansion of the mixing terms  $\exp(it')\Delta_G^2$  to the following transform law [12]:

$$\tilde{I}_R(\omega_0) = \xi_j^2 \sum_k \frac{\Delta_G^{2k}}{k!} \left| \frac{d^k}{d\omega_0^k} [\tilde{\Phi}(\omega_0) - \tilde{\Phi}(\omega_0 - \omega_j)] \right|^2, \quad (21)$$

where the complex refractive index  $\tilde{\Phi}(\omega)$ ,

$$\tilde{\Phi}(\omega) = i \int_0^\infty dt e^{it(\omega - \Omega_{21})} e^{-t^2 \Delta_G^2 / 2} F(t), \quad (22)$$

is determined by the *inhomogeneous* (experimental) absorption spectrum according to Eq. (6).

Analogously, we get for CARS

$$\begin{aligned} \tilde{I}_{\text{CARS}} = & \left| \xi_j^2 \sum_k \frac{\Delta_G^{2k}}{k!} \frac{d^k}{d\omega_0^k} [\tilde{\Phi}(\omega_0) - \tilde{\Phi}(\omega_0 - \omega_j)] \right. \\ & \left. \times \frac{d^k}{d\omega_0^k} [\tilde{\Phi}(\omega_0 + \omega_j) - \tilde{\Phi}(\omega_0)] \right|^2 \end{aligned} \quad (23)$$

and CSRS

$$\begin{aligned} \tilde{I}_{\text{CSRS}} = & \left| \xi_j^2 \sum_k \frac{\Delta_G^{2k}}{k!} \frac{d^k}{d\omega_0^k} [\tilde{\Phi}(\omega_0) - \tilde{\Phi}(\omega_0 - \omega_j)] \right. \\ & \left. \times \frac{d^k}{d\omega_0^k} [\tilde{\Phi}^*(\omega_0 - \omega_j) - \tilde{\Phi}^*(\omega_0 - 2\omega_j)] \right|^2. \end{aligned} \quad (23')$$

In spite of their compact and general form, Eqs. (21) and (23) are not convenient for a practical description of inhomogeneous broadening because of the differentiation operation. They can practically be used only when the inhomogeneous broadening is small in comparison with the homogeneous width. If this condition is not fulfilled, one can use the following numerical procedure:

1) Choose a trial inhomogeneous broadening function  $q(\Omega)$  and find its Fourier transform  $q(t)$ .

2) Calculate the Fourier transform of the experimental absorption spectrum,  $F_{\text{exp}}(t)$ , and then the Fourier transform of the trial homogeneous absorption spectrum

$$F_{\text{tr}}(t) = F_{\text{exp}}(t) q^{-1}(t).$$

3) Calculate the trial excitation profile  $I_{\text{tr}}(\omega_0)$  according to theoretical relations (17) and (18) between the Fourier transform of absorption and the Fourier amplitude of scattering, and its Fourier transform

$$J_{\text{tr}}(\tau) = \int_{-\infty}^{\infty} e^{-ix\tau} I_{\text{tr}}(x) dx.$$

4) Calculate the theoretical excitation profile of scattering

$$I_{\text{theor}}(\omega_0) = (2\pi)^{-1} \int_{-\infty}^{\infty} e^{i\omega_0\tau} Q(\tau) J_{\text{tr}}(\tau) d\tau.$$

In the case of a bad fit all the calculations should be repeated for another  $Q$ . This algorithm needs only the Fourier transform procedures; in principle, it works for an arbitrary shape and width of the inhomogeneous broadening function.

## CONCLUSION

The analysis of coherent Raman scattering excitation profile shape under resonance excitation is a valuable complement to modern resonance Raman spectroscopy. Comparison of CARS and CSRS behaviour allows one to draw conclusions about inhomogeneous broadening. As it was shown, in the homogeneous case the excitation profiles of CARS and CSRS have the same shape/widths. The inhomogeneity of the medium induces a difference between these excitation profiles, while CSRS remains sharper than CARS.

The transform laws which allow to calculate the Raman excitation profiles via the measured absorption spectrum for identical molecules hold for the CARS excitation profile in the inhomogeneous case of a Lorentzian distribution as well; for a Gaussian distribution they are not quite correct any more but nevertheless can practically be used if the inhomogeneous broadening is not large. However, these transform laws do not hold for incoherent REP and CSRS. Here a fitting procedure for calculating real excitation profiles is proposed which allows one to determine the distribution function/width of the inhomogeneous broadening.

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## REFERENCES

1. Tehver, I. Proc. Estonian Acad. Sci. Phys. Math., 1993, 42, 2, 166—172.
2. Champion, P. M., Albrecht, A. C. Ann. Rev. Phys. Chem., 1982, 33, 353—376.
3. Page, J. B. In: Cardona, M., Güntherodt, G. (eds.). Light Scattering in Solids VI. Springer, Berlin—Heidelberg—New York, 1991, 17—72.
4. Hizhnyakov, V., Tehver, I. Phys. Stat. Sol., 1967, 21, 2, 755—768.
5. Hizhnyakov, V. V., Tehver, I. J. Optics Comm., 1980, 32, 3, 419—421; Tehver, I. J. Optics Comm., 1981, 38, 4, 279—283; Hizhnyakov, V., Tehver, I. J. Raman Spectrosc., 1988, 19, 6, 383—388; Hizhnyakov, V., Tehver, I., Zavt, G. J. Raman Spectrosc., 1990, 21, 4, 231—236; Tehver, I. J. Raman Spectrosc., 1991, 22, 4, 211—216.
6. Tonks, D. L., Page, J. B. Chem. Phys. Lett., 1979, 66, 3, 449—453; Chan, C. K., Page, J. B. J. Chem. Phys., 1983, 79, 11, 5234—5250; Brafman, O., Chan, C. K., Khodadoost, B., Page, J. B., Walker, C. T. J. Chem. Phys., 1985, 80, 11, 5406—5417; Chan, C. K., Page, J. B., Tonks, D. L., Brafman, O., Khodadoost, B., Walker, C. T. J. Chem. Phys., 1985, 82, 11, 4813—4830.

7. Stallard, B. R., Callis, P. R., Champion, P. M., Albrecht, A. C. J. Chem. Phys., 1983, 78, 72, 712—722.
8. Desiderio, R. A., Hudson, B. S. Chem. Phys. Lett., 1979, 61, 3, 445—448.
9. Pfeiffer, M., Lau, A., Werncke, W. J. Raman Spectrosc., 1984, 15, 1, 20—28.
10. Lukashin, A. V., Frank-Kamenetskii, M. D. J. Raman Spectrosc., 1982, 12, 3, 234—237.
11. Лэкс М. In: Проблемы физики полупроводников. Москва, 1957, 407—423.
12. Hizhnyakov, V., Tehver, I. J. Raman Spectrosc., 1993 (in press).

## **TRANSFORMSEOSSED NEELDUMISE JA KOHERENTSE RESONANTSE KOMBINATSIOONHAJUMISE VAHEL HOMOGEEN- SES JA MITTEHOMOGEENSES KESKKONNAS**

Imbi TEHVER

Koherentse resonantse (anti-Stokesi ja Stokesi) kombinatsioonhajumise amplituudid/ergastusspektrid on avaldatud optilise neeldumise kaudu, kusjuures on vaadatud, kuidas mõjutab identsete molekulide spektreid ja vastavaid transformseoseid mittehomogeenne keskkond. Siinne töö on loogiline jätk varem väljatöötatud transformmeetodile, mis seostab spontaanse Ramani hajumise ergastusprofiile mõõdetud neeldumisspektriga.

## **СООТНОШЕНИЯ ПРЕОБРАЗОВАНИЯ МЕЖДУ ПОГЛОЩЕНИЕМ И КОГЕРЕНТНЫМ РЕЗОНАНСНЫМ КОМБИНАЦИОННЫМ РАССЕЯНИЕМ В ОДНОРОДНОЙ И НЕОДНОРОДНОЙ СРЕДЕ**

Имби ТЕХВЕР

Амплитуды/спектры возбуждения когерентного (антистоксового и стоксового) комбинационного рассеяния выражены через оптическое поглощение. Рассматривается, как влияет на спектры и соотношения преобразования, полученные в случае идентичных молекул, неоднородная среда. Данная работа является развитием ранее разработанного метода преобразования спонтанного резонансного комбинационного рассеяния на когерентные вынужденные (стоксово и антистоксово) комбинационные рассеяния.