# Raman scattering of light by polaritons coupled to a soft mode of a lithium tantalate crystal

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The temperature dependence of the spectra of Raman scattering (RS) of light by polaritons coupled to a soft mode of a lithium tantalate crystal is investigated. It is shown that the soft mode is by its nature not relaxational, as might follow from investigations of ordinary RS of light by optical phonons, but is a resonant process, albeit strongly overdamped near the phase-transition temperature. At low polariton momenta distinct peaks were observed in the spectra of RS of light at small angles even in the case of appreciable overdamping of the soft mode. The appearance of these peaks has made it possible to measure the temperature dependences of the frequency of the undamped soft mode and of the static dielectric constant at frequencies above the piezoresonance of the crystal.

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## 1. INTRODUCTION

Phase transitions in crystals are accompanied frequently by vanishing of the frequency of one of the lattice modes, called soft mode. Most investigations of the temperature dependence of soft modes are presently carried out by the method of Raman scattering (RS) of light by optical phonons. According to the experimental data, however, in the vicinity of the phase transition the damping of the soft modes increases sharply (the mode becomes overdamped), and the distinct low-frequency peak corresponding to the soft phonon the RS spectra usually have a broad spectrum with a maximum at zero frequency (central peaks) and with a "tail" extending over dozens or even hundreds of $cm^{-1}$ (see, e.g., Refs. 1 and 2). This makes it difficult to obtain information on the soft mode from the spectra of RS by optical phonons.

It is therefore of particular interest to study the spectra of RS of light by polaritons (which appear in small-angle scattering of light in crystals without symmetry center (see, e.g., Ref. 3) and are coupled to the overdamped modes, since the polariton damping decreases with decrease in their momenta, so that peaks corresponding to light scattering by polaritons appear notwithstanding the absence of a distinct peak at the displaced frequency in the spectra of RS by optical phonons.[4,5]

Since the polariton spectra measured in experiments on the spectra of RS of light at small angles are determined by the dispersion relation of an undamped oscillator,[3] investigation of the spectra of RS by polaritons makes it possible to determine the static dielectric constant[1)] $\varepsilon_0$ and the frequency of the soft mode even if the soft mode is strongly overdamped.

This paper is devoted to an experimental investigation of the temperature dependence of the spectra of RS of light by polaritons coupled to an overdamped soft mode of a lithium tantalate crystal. The choice of this crystal was governed by the possibility of realizing synchronism conditions for the observation of light scattering by polaritons of the lower dispersion branch of symmetry $A_1(z)$, which are coupled with the soft mode, and also by the large RS cross section, which facilitates greatly the registration of the scattering spectra.

In addition, the applicability of the soft-mode concept to the interpretation of the ferroelectric phase transition in the lithium tantalate crystal is now extensively discussed in the literature. Thus, for example, by investigating the temperature dependence of the spectra of RS by optical phonons, Johnston and Kaminov[6] found that the frequency of one of the transverse optical (*TO*) oscillations decreases ("softens") to $\sim 100\ cm^{-1}$ as the crystal approaches the Curie temperature $T_c$ [in proportion to $(T - T_c)^{1/2}$], and reached the conclusion that the phase transition is of second order and of the displacive type. The soft mode was overdamped and its frequency was determined from the position of the "center of gravity" of the observed spectral distribution, a conclusion that is generally speaking incorrect.[2] Ivanova *et al.*,[7] however, observed no anomalous frequency displacement of any of the maxima in the RS spectrum as the temperature of the ferroelectric phase transition temperature was approached, although the general form of the spectrum changed quite substantially. It was concluded on this basis that the soft-mode concept is not aplicable.[7] That there is no soft mode is indicated also in some other papers on RS,[8,9] although the data on IR reflection are well explained within the framework of the soft-mode concept.[10]

## 2. SPECTRAL DISTRIBUTION OF THE INTENSITY OF RS BY POLARITONS COUPLED WITH DAMPED OPTICAL PHONONS

We consider the spectral distribution of the intensity $J(\nu, k)$ of RS of light by polaritons at different values of the damping of optical phonons transformed into polaritons at low wave-vector values $k$. We start for simplicity from the oscillator model and assume the following:

(a) $\varepsilon_0 \gg \tilde{\varepsilon}_\infty$, where $\varepsilon_0$ is the static dielectric constant defined above and $\tilde{\varepsilon}_\infty$ is the "background" dielectric constant determined by the contributions of all the (electronic and lattice) excitations of the crystal, with the exception of the oscillator considered. This assumption is valid when the

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main contribution to $\varepsilon_0$ is determined by the considered oscillator that describes, say, a soft mode in ferroelectrics.

(b) $c^2k^2/\omega^2 \gg \tilde{\varepsilon}_\infty$, which is valid when the lower polariton branch is considered. Here $c$ is the speed of light in vacuum and $\omega = 2\pi c\nu$ is the cyclic frequency. In this case the function $J(\nu,k)$ (without allowance for the dispersion of the nonlinear susceptibility in the high-temperature limit) can be represented in the form[5]

$$J(\nu,k) \sim (n+1)\,\mathrm{Im}\,\chi(\nu,k) \sim \frac{\nu_p^2(k)}{\nu_0^2}\,\frac{\Gamma_p(k)}{[\nu_p^2(k)-\nu^2]^2+\nu^2\Gamma_p^2(k)} \tag{1}$$

$$\nu_p^2(k) = \frac{\nu_0^2}{1+Q^{-2}}, \quad \Gamma_p(k) = \frac{\Gamma}{1+Q^{-2}}, \quad Q = \frac{k}{2\pi\nu_0\sqrt{\varepsilon_0}}, \tag{2}$$

$\nu_0$ and $\Gamma$ are the frequency and damping of the optical phonon (of the soft mode), $\nu_p(k)$ and $\Gamma_p(k)$ are the frequency and damping of the polaritons, and $n$ is the Bose factor.

Figure 1 shows the spectral distributions of the intensities $J(\nu, k)$ of the RS of light by polaritons at different momentum values, for three ratios of the optical-phonon frequency $\nu_0$ to its damping $\Gamma$. The frequencies and the damping were chosen such that the changes of the spectra corresponded qualitatively to the spectra of RS by polaritons coupled with the soft mode, i.e., the damping constant $\Gamma$ increases with decreasing phonon frequency $\nu_0$ (cf. *a*, *b*, and *c*).

It can be seen from Fig. 1a (case $\Gamma \ll \nu_0$) that the line of the RS of light by polariton becomes narrower with decreasing polariton momentum, and the positions of the maxima of these lines are determined with good accuracy by the dispersion relation (2), which does not depend on the damping constant (the dashed $\nu_p$ curve).

At $\Gamma = \nu_0$ (Fig. 1b) in the region of large $k$ ($Q > 1$) the spectra become diffuse, and the positions of their maxima can hardly be determined in experiment, since the diffuse character of the spectra is also aggravated by the "instrumental wing" due to the scattering by the undisplaced component. It should be noted that the positions of the maxima are in this case no longer determined by the polariton dispersion law (2), since it follows from (1) that the polariton frequency $\nu_p(k)$ corresponds to the maximum of the function $S = \omega\,\mathrm{Im}\chi(\nu, k)$.

In the region of small $k$ ($Q \lesssim 0.5$), however, distinct albeit asymmetric peaks appear in the scattering spectra. The described features of the spectra manifest themselves particularly strongly at large and small $k$ in the case when the damping is even larger (Fig. 1c). At values $Q \gtrsim 0.5$ the spectra merge with the spectral peak, which can no longer be distinguished at large $Q$ from the peaks produced, e.g., in the relaxation process. At small $k$ ($Q < 0.5$) distinct peaks appear in the spectra, and by measuring their positions (in $k$, $\omega$ space) we can obtain the polariton dispersion, and hence also the values of $\varepsilon_0$ and $\nu_0$.

Indeed, we rewrite the dispersion relation (2) in the form

$$k^2/4\pi^2\nu_p^2(k) = \varepsilon_0\nu_0^2/[\nu_0^2-\nu_p^2(k)]. \tag{3}$$

It is seen therefore that the slope of the dispersion curve $\nu_p(k)$ as $\nu_p \to 0$ is determined by the value of $\varepsilon_0$. Consequently $\varepsilon_0$ can be easily determined from the polariton scattering spectra at low values of the polaritons wave vectors (frequencies) even when the phonon damping exceeds substantially their frequency (Fig. 1c). We note that $\varepsilon_0$ is determined by the slope of the dispersion curve $\nu_p(k)$ independently of the approximations made in the derivation of relations (1)–(3), since the exact expression for $k(\nu_p)$ (Ref. 3)

$$\frac{k^2}{4\pi^2\nu_p^2} = \varepsilon_\infty + \sum_i \frac{f_i\nu_i^2}{\nu_i^2-\nu_p^2}$$

yields as $\nu_p \to 0$

$$\frac{k^2}{4\pi^2\nu_p^2} = \varepsilon_\infty + \sum_i f_i = \varepsilon_0, \tag{4}$$

where $f_i$ is the oscillator strength of an optical phonon of frequency $\nu_i$; the summation is over all the dipole-active optical phonons of the considered symmetry type.

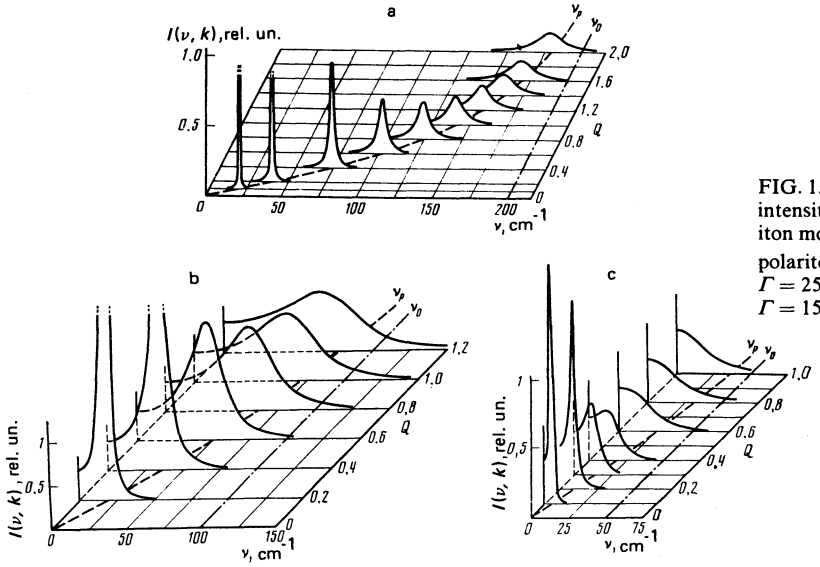


FIG. 1. Calculated dependences of the spectral distribution of the intensity of RS of light by polaritons at various values of the polariton momenta $k$; $\nu_0$ is the optical-phonon frequency, $\nu_p$ is the polariton dispersion curve, $Q = k/2\pi\nu_0\sqrt{\varepsilon_0}$, a) $\nu_0 = 200\ \mathrm{cm}^{-1}$, $\Gamma = 25\ \mathrm{cm}^{-1}$; b) $\nu_0 = 100\ \mathrm{cm}^{-1}$, $\Gamma = 100\ \mathrm{cm}^{-1}$; c) $\nu_0 = 50\ \mathrm{cm}^{-1}$, $\Gamma = 150\ \mathrm{cm}^{-1}$.

If it is assumed that the $\varepsilon_0(T)$ temperature dependence is determined only by the change of the soft-mode frequency with temperature,[11] we can find from the generalized Lyddane-Sachs-Teller relation that

$$\varepsilon_0(T)=\frac{1}{\nu_0^2(T)}\left(\varepsilon_\infty\nu_{0,L}^2\prod_{i\neq 0}\frac{\nu_{i,L}^2}{\nu_i^2}\right)=\frac{\alpha}{\nu_0^2(T)}, \qquad (5)$$

where $\nu_{i,L}$ is the frequency of the longitudinal component of the $\nu_i$ oscillations, and $\alpha$ is independent of temperature by virtue of the assumption made above and of the experimental fact that $\nu_{0,L}$ is practically independent of temperature. Thus, for example, by determining $\alpha$ at room temperature from measurements of the polariton spectra, or by calculating it from data on RS scattering by optical phonons in accordance with (5), we can obtain the temperature dependence of the soft-mode frequency from the temperature measurements of the RS of light by polaritons even when the soft-mode damping $\Gamma$ exceeds substantially its frequency $\nu_0$.

### 3. EXPERIMENTAL TECHNIQUE

We have investigated in the experiment the temperature dependence of RS of light by polaritons using a photographic technique (wherein the spectrograph slit is placed in the focal plane of the lens that gathers the scattered light). This technique yields the frequency-angle spectra (the dependence of the frequency $\nu$ of the RS shift on the scattering angle $\varphi$).[3] The spectra were photographed with an ISP-51 spectrograph at using a slit $\lesssim 15\ \text{cm}^{-1}$ wide. The scattered-light spectra were excited by a single-frequency single-mode argon laser at a wavelength 5145 Å, whose frequency was tuned with a stabilized Fabry-Perot interferometer, placed in the cavity, to the absorption line of molecular iodine, which prevented entry of the exciting radiation into the spectrograph. The power of the single-frequency laser was about 300 mW. The experiments were performed at scattering geometry $y(z, z)\, y + \Delta x$ at which, according to the selection rule, only polaritons of symmetry $A_1(z)$ appear in the scattering spectra of the lithium tantalate crystal. This geometry makes it also possible to observe the entire lower polariton branch coupled with the soft mode. The lithium tantalate crystal was placed in an opetical thermostat in which the crystal temperature could be regulated in the range from room temperature to 650 °C accurate to $\pm$ 5 °C. The crystal measured approximately 15×15×15 mm.

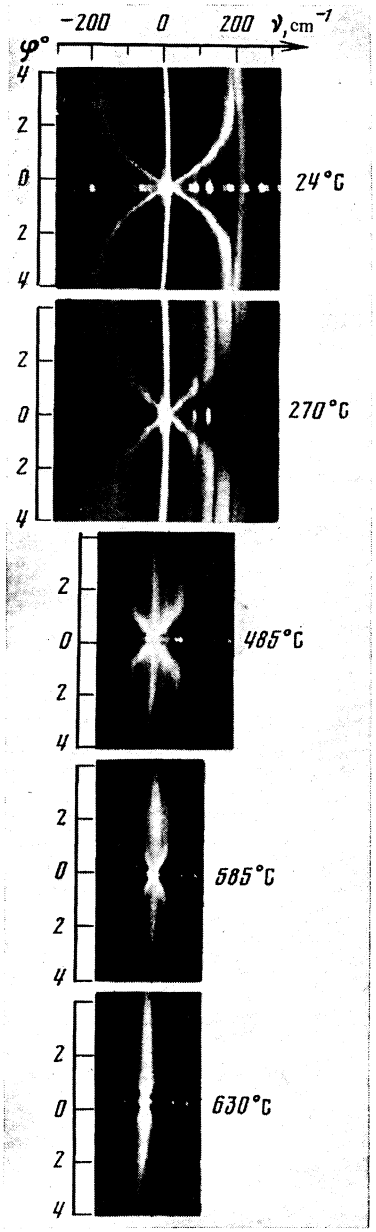


FIG. 2. Frequency-angle spectra of RS of light by polaritons of the lower branch, which are coupled with the soft mode of symmetry $A_1(z)$ of the lithium-tantalate crystal. $\nu$ is the frequency shift of RS and $\varphi$ is the scattering angle reckoned inside the crystal. The spectra were obtained at crystal temperatures 24, 270, 485, 858, and 630 °C.

### 4. EXPERIMENTAL RESULTS

Figure 2 shows the frequency-angle spectra of RS of light by polaritons of the lower dispersion branch, obtained for different temperatures of the lithium tantalate crystal. The vertical dark bands on the spectrograms are due to absorption of the scattered light by the lines of the molecular iodine used to absorb the exciting radiation (these bands are most clearly seen in the scattering spectrum obtained at a crystal temperature 270 °C). Negative frequency shifts correspond to anti-Stokes scattering.

The frequency-angle scattering spectrum obtained at a crystal temperature 24 °C corresponds to the condition $\Gamma \ll \nu_0$, and the intensity distribution corresponds in this case to the model spectrum shown in Fig. 1a. It can be seen from Fig. 2 that the frequency $\nu_0$ of the optical phonon decreases and its damping increases with increasing crystal temperature. At 270 °C the optical-phonon line width becomes comparable with its frequency ($2\Gamma \sim \nu_0$). At low scattering angles $\varphi$ (at low values of the polariton momenta), however, the polariton branch remains quite narrow. With further rise of temperature a substantial modification of the frequency-angle spectra takes place. The spectrum becomes diffuse, but has in the region of small scattering angles ($\varphi \lesssim 1°$) a sharp boundary determined by the polariton maxima in the spec-

tral distribution of the intensity of the RS by the polaritons. The character of the observed spectra is in good qualitative agreement with the model spectra shown in Figs. 1b and 1c, and corresponds to the case $\Gamma > \nu_0$. We note, however, that at a crystal temperature 630 °C the intensity "dip" between the Stokes and anti-Stokes peaks of the light scattering by polaritons, a peak that follows from Fig. 1c, does not appear in the scattering spectrum. This is due to the very small shift of the frequencies of the light scattered by the polaritons (owing to the low frequency of the soft mode at this temperature), and hence also to the small frequency spacing between the Stokes and anti-Stokes peaks in the polaritons scattering. Nonetheless, this spectrum shows distinctly the angle-dependent boundary of the diffuse spectrum in the region of small scattering angles $\varphi$, in agreement with the model spectra of Fig. 1c. It should also be noted that at high crystal temperatures (500 °C) the diffuse spectrum with center at $\nu = 0$ cm$^{-1}$ first broadens with increasing scattering angle, follows the polariton maxima, reaches a certain maximum width, and then "contracts" gradually to zero frequency. This is caused by the strong overdamping of the soft mode at temperatures close to the phase-transition temperature of the lithium tantalate crystal.

The spectra obtained make it possible, as indicated above, to determine the static dielectric constant $\varepsilon_0$ at various crystal temperatures. According to (2) and (4) the static dielectric constant is determined from the slope (asymptote) of the lower polariton dispersion branch $2\pi\sqrt{\varepsilon_0} = (dk/d\nu_p)_{\nu_p=0}$ or directly from the frequency-angle spectra of the RS of light by polaritons:

$$\varepsilon_0 = \left[n_l + \nu_l \left(\frac{\partial n}{\partial \nu}\right)_{\nu=\nu_l}\right]^2 + (\nu_l n_l)^2 \left(\frac{\partial \varphi}{\partial \nu_p}\right)^2_{\nu_p=0}, \qquad (6)$$

where $n_l$ is the refractive index of the crystal at the exciting-radiation wavelength.[2)]

The value of $(\partial\varphi/\partial\nu_p)^2_{\nu_p=0}$ determined in experiment by measuring the frequency-angle spectrum of the RS of light by polaritons of a lithium-tantalate crystal at 24 °C amounts to $2.1\times10^{-8}$ rad$^2$·cm$^2$. Using the refractive index data of Ref. 12 we obtain from (6) $\varepsilon_0 = 45 \pm 5$. This results agrees well with the data obtained from the spectra of RS by polaritons[13] ($\varepsilon_0 = 44$) and from direct high-frequency measurements[14]: $\varepsilon_0 = 42.2$ at 0.1 GHz and $\varepsilon_0 = 41.4$ at 1 GHz. A somewhat higher value $\varepsilon_0 = 48$ is obtained by calculation in accord with the Lyddane-Sachs-Teller formula using data on RS of light by optical phonons. In the calculation of $\varepsilon_0$ in Ref. 13, however, they used for $\varepsilon_\infty$ a value 4.9, which is the refractive index of lithium tantalate crystals at a wavelength 0.5 $\mu$m. It must be noted that some leeway in the choice of $\varepsilon_\infty$ (there is, of course, no rigorous criterion for the choice of $\varepsilon_\infty$) leads often to a noticeable deviation of the calculated dispersion of the upper polariton branch from experiment (see, e.g., Refs. 15 and 16). It is most reasonable to choose for $\varepsilon_\infty$ values at wavelengths in the transparency region of the crystal, where the dispersion of the refractive index is a minimum. Assuming for lithium tantalate $\varepsilon_\infty = \varepsilon$ ($\lambda = 1.8$ $\mu$m) = 4.48 (Ref. 12), we find from data on RS of light by optical phonons[13] that $\varepsilon_0 = 43.9$. This result agrees with good accuracy with the measured values cited above.

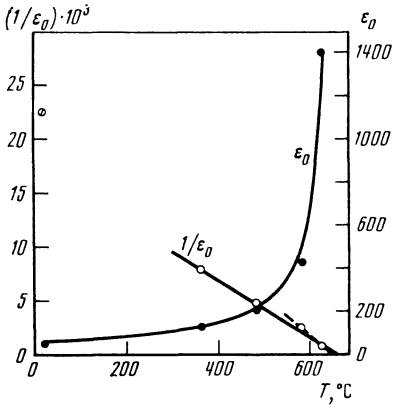


FIG. 3. Temperature dependence of static dielectric constant $\varepsilon_0(T)$. Points—measurement results. Solid curve—calculation using linear approximation of the temperature dependence of the reciprocal of the dielectric constant $1/\varepsilon_0$, obtained using four experimental points. The dashed straight line is drawn through the two points obtained at the highest temperatures of the crystal.

The static dielectric constants obtained by measuring the spectra of the RS of light by polaritons at various lithium-tantalate crystal temperatures are represented by the dark points in Fig. 3. We note that at a crystal temperature 630 °C, when the soft mode becomes strongly overdamped and its frequency is considerably lowered, it is difficult to discern on the spectrogram (see Fig. 2) the distinct peak due to light scattering by polaritons. We have therefore measured $\varepsilon_0$ by determining the asymptote of the "abrupt boundary" of the diffuse spectrum on the side of small scattering angles (see also Fig. 2c). The error in the measurement of $\varepsilon_0$ is in this case approximately 15%. The light points in Fig. 3 show also the values of the reciprocal values of the dielectric constant. It can be seen from the figure that at high crystal temperatures the deviation of the experimental from the linearity (solid straight line) calculated by least squares using four points, exceeds the measurement error. The Curie-Weiss constant and the phase-transition temperature were therefore calculated using only two values of $\varepsilon_0$ (through which the dashed line was drawn) measured at temperatures[3)] 585 and 630 °C:

$$\varepsilon_0(T) = 2.8\cdot10^4(°\,\mathrm{C})/(T_0 - T), \qquad (7)$$

where $T_0 = 650$ °C. It should be noted that the lithium-tantalate crystal phase-transition temperature can range, depending on the growth conditions, from 510 to 690 °C (Ref. 2). Thus, for example, $T_0 = 676$ °C for a stoichiometric crystal.[2]

Since the polariton frequency is determined by the dispersion relation (2), i.e., in terms of the frequency of the undamped optical phonon, we can determine from the obtained frequency-angle spectra of RS by polaritons also the frequency of the soft mode at various crystal temperatures. Using (5) and (6) we obtain for the temperature dependence of the soft-mode frequency

$$\nu_0^{-2}(T)=\frac{\varepsilon_0(T)}{\alpha}$$

$$=\alpha^{-1}\left\{\left[n_l+\nu_l\left(\frac{\partial n}{\partial \nu}\right)_{\nu=\nu_l}\right]^2+(\nu_l n_l)^2\left(\frac{\partial \varphi(T)}{\partial \nu_p}\right)^2_{\nu_p=0}\right\}.$$

The constant $\alpha\approx 1.8\times 10^6$ cm$^{-2}$ was in fact determined from the frequency-angle scattering spectra of the crystal at room temperature, since under these conditions $\nu_0$ can be measured directly alongside $\varepsilon_0$. The resultant frequencies of the soft mode[4] were: $\nu_0(364\ ^\circ\text{C}) = 120\ \text{cm}^{-1}$, $\nu_0(485\ ^\circ\text{C} = 90\ \text{cm}^{-1}$, $\nu_0(580\ ^\circ\text{C}) = 65\ \text{cm}^{-1}$, and $\nu_0(630\ ^\circ\text{C}) = 36\ \text{cm}^{-1}$.

## 5. DISCUSSION OF RESULTS AND CONCLUSIONS

Investigations of the temperature dependence of the spectra of RS of light by polaritons coupled with the soft mode of the lithium-tantalate crystal have shown that polariton peaks are observed in the scattering at low values of the wave vectors (at small scattering angles) even in the case of strongly overdamped optical phonons. The appearance of these peaks has made it possible to measure the frequency dependences of the soft-mode frequency without damping and of the static dielectric constant at frequencies above the piezoresonances of the crystal, something impossible before by using ordinary RS of light by optical phonons, owing to the strong overdamping of the soft mode. Thus, the soft mode in lithium tantalum is by its nature not relaxational, as would follow from Refs. 7–9, but resonant, albeit strongly overdamped. This conclusion is in accord with investigations of IR reflection spectra.[10]

It should be noted that the character of the experimentally obtained spectra is qualitatively in good agreement with the spectra calculated on the basis of the simplest model of a harmonic oscillator with strong damping. For a quantitative comparison of the spectra distribution of the scattering intensities, however, further development of the theory of RS of light by polaritons of overdamped optical phonons is necessary, with account taken of the frequency and temperature dependences of the damping constant.[5] It must also be taken into account here the dispersion of the nonlinear susceptibility that describes the RS of light by polaritons (see, e.g., Ref. 3). Allowance for the dispersion of the nonlinear susceptibility can lead in this case also to an additional change in the line shape of the light scattered by the polaritons.

The possible mechanisms of the frequency and temperature dependences of the damping constants of soft modes are discussed in reviews by Scott[18] and Ginzburg *et al.*[19] One of the undisputed mechanisms of this dependence in the case of lithium tantalate crystals is the interaction of the soft mode with the second-order phonon spectrum.[20] We call attention to the fact that, for example in the spectrum obtained at 585 °C (Fig. 2), a scattering minimum that does not follow from the model spectra of Fig. 1 can be seen between the polariton peak and zero frequency (at constant $k$ or $\varphi$). It appears that such a singularity can appear when account is taken of the frequency dependence of the damping constant.

In conclusion, we are deeply grateful to A. M. Prokhorov and P. P. Pashinin for support.

[1)]At frequencies much higher than that of piezoresonance but lower than the soft-mode frequency of the investigated crystal.

[2)]This relation follows directly from the energy and momentum conservation laws for the elementary acto of RS of light by polaritons at small scattering angles $\varphi$ and in the approximation $n_s = n_l - (\partial n/\partial \nu)_{\nu=\nu_l}\nu_p$, where $n_s$ is the refractive index at the scattered-light frequency.

[3)]We assume here that the temperature dependence of the dielectric constant is described by the Curie-Weiss law, which usually is well satisfied at $(T_0 - T)/T_0 \lesssim 0.1$.

[4)]The change of the refractive indices $n_l(T)$ in the considered temperature interval is[17] $\lesssim 3\%$ and can be neglected.

[5)]We note that the exact expression obtained so far for the intensities of RS of light by polaritons were derived in the approximation $\Gamma \ll \nu_0$ and cannot be used by formally substituting in them the frequency- and temperature-dependent damping.


[1]R. Blinc and B. Žeks, Soft Modes in Ferroelectrics and Antiferroelectrics, North-Holland, 1974.
[2]M. E. Lines and A. Glass, Principles and Applications of Ferroelectrics & Related Materials, Oxford U. P., 1977.
[3]U. N. Polivanov, Usp. Fiz. Nauk **126**, 185 (1978) [Sov. Phys. Usp. **21**, 805 (1978)].
[4]L. Laughman, L. W. Davis, and T. Nakamura, Phys. Rev. **B6**, 3322 (1972).
[5]T. Nakamura and Ya. Tominaga, Izv. AN SSSR, Ser. Fiz. **41**, 579 (1977).
[6]W. D. Johston, Jr. and I. P. Maminov, Phys. Rev. **168**, 1045 (1968).
[7]S. V. Ivanova, V. S. Gorelik, and B. A. Strukov, FIAN Preprint No. 134, 1978.
[8]A. F. Penna, A. Chaves, and S. P. S. Porto, Sol. St. Commun. **19**, 491 (1976).
[9]A. F. Penna, S. P. S. Porto, and E. Wiener-Avnear, Sol. St. Commun. **23**, 377 (1976).
[10]J. L. Servoin and F. Gervais, Sol. St. Commun. **31**, 387 (1969).
[11]J. F. Scott, P. A. Fleury, and J. W. Worlock, Phys. Rev. **177**, 1288 (1969).
[12]W. L. Bond, J. Appl. Phys. **36**, 1674 (1965).
[13]A. F. Penna, A. Chaves, P. da R. Andrade, and S. P. S. Porto, Phys. Rev. **B13**, 4907 (1976).
[14]J. R. Teque, R. R. Rice, and R. Gerson, J. Appl. Phys. **46**, 2864 (1975).
[15]L. A. Kulevsky, Ya. N. Polivanov, and S. N. Pluektov, J. Raman Spectrosc. **3**, 239 (1975).
[16]L. I. Kuznetsova, L. A. Kulevskii, Yu. N. Polivanov, and K. A. Prokhorov, Kvant. Elektron. (Moscow) **2**, 2095 (1975) [Sov. J. Quantum Electron. **5**, 1146 (1975)].
[17]H. Iwasaki, T. Yamada, N. Niizeki, H. Toyoda, and J. Kubota, Jpn. J. Appl. Phys. **7**, 185 (1968).
[18]J. F. Scott, Rev. Mod. Phys. **46**, 83 (1974).
[19]V. L. Ginzburg, A. P. Levanyuk, and A. A. Sobyanin, Usp. Fiz. Nauk **130**, 615 (1980) [Phys. Repts. **S7** (3), 152 (1980)].
[20]Yu. N. Polivanov and S. N. Poluektov, Fiz. Tverd. Tela (Leningrad) **20**, 3482 (1978) [Sov. Phys. Solid State **20**, 2014 (1978)].


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