SPECTRUM OF SURFACE POLARITONS ON GaSe CRYSTALS

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The spectrum of surface vibrations on GaSe exited by the optical method of attenuated total reflection (ATR) has been studied theoretically. The ATR spectra have been calculated for the geometry when the normal to the crystal surface is perpendicular to the cleavage plane. The frequency-wavevector dispersion determined by positions of the ATR spectrum minima at various angles of incident light within frequency range $214<\nu<254$ cm^{-1} consists of two branches. The lower branch ($\nu<245$ cm^{-1}) corresponds to the surface mode of type I (real mode) or type II (virtual mode) depending on the wavevector value. The upper branch ($\nu>245$ cm^{-1}) corresponds to the type II surface mode. The results of the calculation have been compared with experimental data obtained by other methods. The calculated dispersion relation at small and large values of the wavevector agrees with experimental data of infrared and Raman spectroscopy.

1. Introduction

In papers [1,2], spectra of the Raman scattering (RS) in GaSe films were investigated. In high-frequency region ($240<\nu<320$cm^{-1}), the RS peaks were observed at the frequencies of ν_1=245cm^{-1}, ν_2=254cm^{-1} and ν_3=309cm^{-1}. For the peaks at ν_2 and ν_3, the RS intensity decreases with the film thickness, whereas the intensity of the peak at ν_1 remains almost invariable. It was explained by a great contribution of surface vibrational states to the RS cross-section at the frequency of 245cm^{-1}. In papers [3,4], the peak near of 245cm^{-1} in the spectra of infrared absorption was observed owing to thin metal (Al) layer evaporated on GaSe films. Generally, the study of surface modes by Raman and infrared spectroscopy methods is difficult because of weak intensity of the scattering [5]. A sensitive and relatively easy technique to study the surface phonon-polaritons on crystals is the method of attenuated total reflection (ATR). By the ATR method a measurement is made of the reflection spectrum of electromagnetic wave incident on the boundary surface of two media (prism-spacing layer) which is a plane of total internal reflection. The presence of a crystal at a distance from this plane involves a violation of the total reflection condition. As a result, the reflection spectrum has sharply defined minima which correspond to the light absorption due to surface phonon-polaritons [5].

For the present, the ATR spectroscopy of surface modes in GaSe has not been done. Theoretical study made in the present paper shows that the ATR spectroscopy method can substantially supplement the results of papers [1-4]. In GaSe, the ATR method will also allow observations of the surface modes of type II (so-called 'virtual excitation' surface polaritons having no analogue in isotropic crystals).

2. Reflection Spectra

The reflection coefficient of light R has been calculated by the method used in paper [6] for uniaxial crystals in accordance with conventional experimental set-up. The prism region and the region of the crystal involved are assumed to be semi-infinite along the z-axis (this axis is perpendicular to the crystal surface and the prism base). The x-axis is brought into coincidence with the plane of light incidence. The case of p-polarized light (i.e. when light is polarized in the plane of incidence) has been considered because the interaction with surface phonons does not occur in s-polarization [5]. The light absorption only by the crystal has been taken into account.

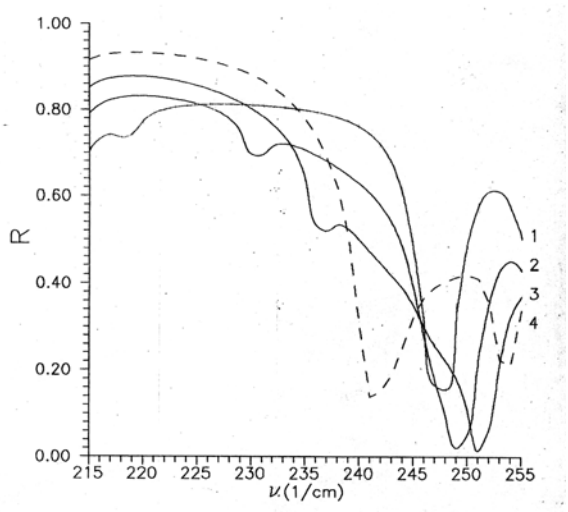

Fig.1. The frequency dependence of the light reflection coefficient R when the light incidence angle is equal to (1) 20^0, (2) 23^0, (3) 25^0 and (4) 28^0.

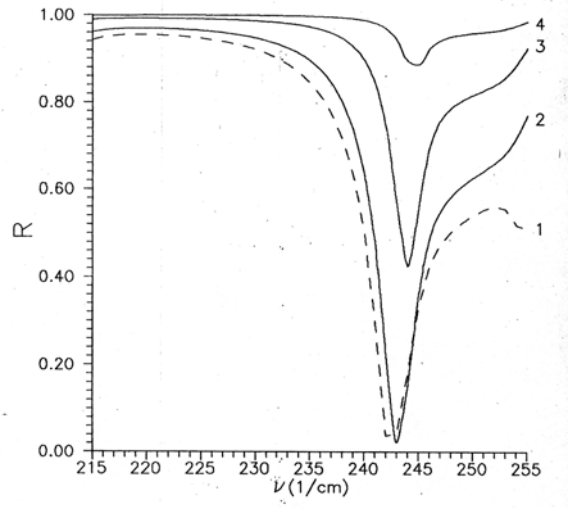

Fig.2. The frequency dependence of the light reflection coefficient R when the light incidence angle is equal to (1) 30^0, (2) 32^0, (3) 40^0 and (4) 60^0.

An expression for R has been found from the solution of a system of equations for the electric field vectors of incident and reflected waves on the prism-spacing and spacing-crystal boundaries. The expression for R has very comber some form (it is similar to that obtained in paper [6]). Numerical

calculations of the ATR spectra have been made for the most conventional geometry of experiments when the normal to the crystal surface is perpendicular to the cleavage plane. It is known that the GaSe crystals are easily cleaved parallel to the layers (the *C* axis of the highest symmetry is perpendicular to the cleavage plane). For calculations, the values of the transverse ν_{TO} and longitudinal ν_{LO} optical phonon frequencies, the damping constant γ and the high-frequency dielectric constant ε_∞ of GaSe given in Table 1 have been used.

Table 1.

Values of the transverse ν_{TO} and longitudinal ν_{LO} optical phonon frequencies, the damping constant γ and the high-frequency dielectric constant ε_∞ for the directions parallel and perpendicular to the optical axis *C* of the GaSe crystals [7].

	$\|C$	$\perp C$
ν_{TO} (cm^{-1})	237	213.5
ν_{LO} (cm^{-1})	245.5	254.7
γ (cm^{-1})	2.8	3
ε_∞	5.76	7.44

A thickness of air spacing between the prism and the crystal surface is taken to be 2.5 μm. The dependence $R(\nu)$ has been calculated for various angles of light incidence φ. In experiments, a set of silicon prisms (ε_{Si} =11.6) with different base angles is often used to obtain the ATR spectra over a range from 18 to 60° [8]. The calculated reflection spectra are shown in figures 1 to 2.

3. Discussion

In figure 1, it is seen that the reflection spectrum has two minima if φ<30°. When the angle of light incidence φ increases, then the depth of minima changes, and their position shifts towards the region of higher frequencies. At φ=28° these minima have comparable depth. If φ>30°, the $R(\nu)$ dependence has one minimum (fig. 3).

Points on figure 3 corresponds to the position of the $R(\nu)$ minima depending upon the reduced wavevector of surface polariton $\kappa=q_x/2\pi\nu$. The relation between κ and the angle of light incidence φ is expressed by the formula

$$\kappa = \sqrt{\varepsilon_{Si}}\, \sin\varphi$$

In figure 3, it is seen that there are two dispersion branches. If κ<1.5, the lower branch corresponds to a surface mode of type II (virtual mode) because within the frequency range 214< ν<237 cm^{-1} the dielectric constant component ε_z is positive and greater that κ^2 (the *z*-axis is parallel to the *C* axis). If κ>1.5, the lower branch corresponds to a surface mode of type I (it is a real mode because ε_z<0): at κ=2 its frequency reaches the value of 244cm^{-1} (the further change of the frequency is small). The upper branch corresponds to the type II surface mode ($\varepsilon_z<\kappa^2$ within the frequency range from 246 to 254cm^{-1}): if κ>1.6, the vibrations corresponding to this branch vanish (according to [5], the point where $\varepsilon_z=\kappa^2$ and $\varepsilon_x=0$ is a stop point for the vibrations). The region of small wavevectors (κ<1) corresponds to the radiative surface modes studied in papers [3,4]. In figure 3, the frequency value of 245cm^{-1} is shown by dashed line. When κ<1, the dashed line corresponds to the film mode observed in paper [3]. At κ>>1 (in papers [1,2] the measurements were made for this region), the dashed line corresponds to the peak ν_1 observed in the RS spectra. From figure 3, it follows that the calculated dispersion curve agrees well with the experimental data available at small and great values of κ. The present paper to be a stimulus for setting up experiments on GaSe by the ATR metod. Results of such experiments will make the comparison in intermediate region of the wavevector values possible.

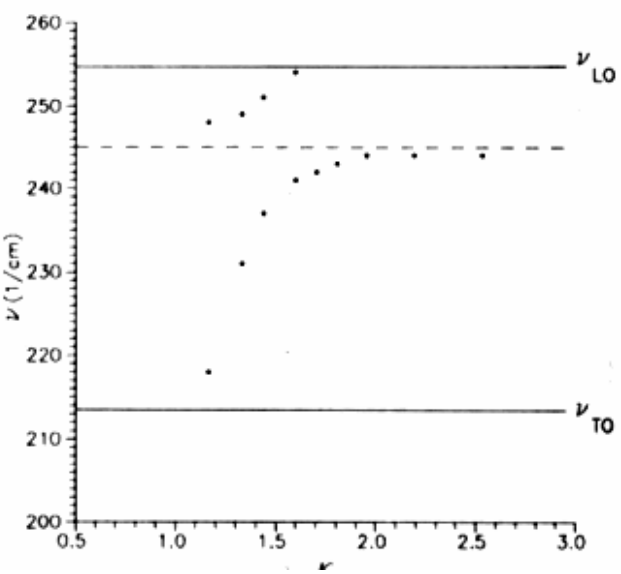

Fig. 3. The dispersion curve of surface polaritons. The points correspond to minima of the $R(\nu)$ dependence given in figures 1 and 2. Solid lines show the frequencies of transverse ν_{TO} and longitudinal ν_{LO} optical phonons in the directions parallel and perpendicular to the optical axis *C* of the GaSe crystals. The dashed line corresponds to the experimental data available at small (κ<1) and great (κ>>1) values of the reduced wavevector κ.

4. Conclusion

The spectrum of surface vibrations on GaSe excited by the optical method of attenuated total reflection has been calculated. It is shown that the dispersion curve has two branches. One of the branches corresponds to the surface mode of type II (virtual mode). The other branch corresponds to the surface mode of type II or to the surface mode of type I (real mode): it depends on the wavevector value of surface polaritons. Results of the calculation agree well with experimental data obtained by other optical methods.

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GaSe KRİSTALINDA SƏTHİ POLYARİTONLARIN SPEKTRİ

Laylı GaSe kristallarında işığın tam daxili qayıtmasını pozma metodu ilə həyəcanlandırılmış səth rəqslərinin spektri nəzəri cəhətdən tədqiq olunub. Spektrin hesablanması, kristal səthinin parçalanma müstəvisinə təsadüf etdiyi hal üçün aparılmışdır. İşığın düşmə bucağının müxtəlif qiymətlərində qayıtma spektrində müşahidə olunan minimumların vəziyyətinə görə müəyyən edilmiş dispersiya asılılığı 214<v254 sm-1 tezlik diapazonunda iki əyridən ibarətdir. Alt əyri (v<245 sm-1), dalğa vektorundan asılı olaraq ya I tip (real), ya da ki, II tip (virtual) səth modasına müvafiqdir. Üst əyri (v>245sm-1), II tip səth modasına aiddir. Alınan nəticələr başqa metodlarla aparılmış təcrübələrin nəticələri ilə müqayisə edilmişdir.

Надир Б. Мустафаев

СПЕКТР ПОВЕРХНОСТНЫХ ПОЛЯРИТОНОВ В КРИСТАЛЛАХ GaSe

Теоретически исследован спектр поверхностных колебаний в GaSe, возбуждаемых оптическим методом нарушенного полного внутреннего отражения (НПВО). Расчет спектров НПВО проведен для геометрии, когда нормаль к поверхности кристалла перпендикулярна плоскости скола. Дисперсионная зависимость, определенная по положению минимумов спектра НПВО при различных углах падения света, в диапазоне частот $214<\nu<254$ см$^{-1}$ состоит из двух ветвей. Нижняя ветвь ($\nu<245$см$^{-1}$) соответствует, в зависимости от величины волнового вектора, поверхностной моде типа I (реальная мода), или же типа II (виртуальная мода). Верхняя ветвь ($\nu>245$ см$^{-1}$) относится к поверхностной моде типа II. Результаты расчета сопоставлены с экспериментальными данными, полученными другими методами.

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