Evidence for exceptional bulk waves on (110) and (111) surfaces of GaAs from Brillouin spectroscopy

1994 J. Phys.: Condens. Matter 6 3359

(http://iopscience.iop.org/0953-8984/6/18/012)

J. Phys.: Condens. Matter 6 (1994) 3359–3368. Printed in the UK

Evidence for exceptional bulk waves on (110) and (111) surfaces of GaAs from Brillouin spectroscopy

V V Aleksandrov and A V Gladkevitch

Chair for Crystallophysics, Physics Department, Moscow State University, Moscow 117234, Russia

Received 14 February 1994

Abstract. Observation of light scattered from (110) and (111) faces of a GaAs crystal reveals a new type of Brillouin line of surface origin when incident light is polarized normally to the plane of light incidence. These new lines are attributed to the presence of equilibria exceptional waves, or bulk shear horizontal acoustic modes satisfying free boundary conditions, and propagating along $[1\bar{1}0]$ and $[1\bar{2}1]$ crystallographic directions, respectively. Reasonable agreement between calculated and measured exceptional wave velocity values is obtained.

1. Introduction

The Brillouin spectroscopy method is known to be an effective and non-destructive tool for the examination of the equilibrium distributions of acoustic phonons at the surfaces of opaque solids [1, 2]. Recent investigations have enabled one to not only describe the details of the propagation of surface waves of the Rayleigh type (RWs), but also to analyse the intensive surface phonon components corresponding to the leaky modes (LMs) of transverse character, and those of the longitudinal ones known as longitudinal resonances (LRs) [3–11].

It should be mentioned that the surface equations of motion problem, in addition to the solutions of Rayleigh mode (RM) or LM type, also have one partial homogeneous plane bulk shear wave solution satisfying boundary conditions, called an exceptional wave (EW) [12–14]. The polarization direction of the EW lies in the boundary surface. It exists at certain azimuthal propagation directions at which the RW, consisting generally of three partial modes, degenerates into two partial modes with displacements in the sagittal plane.

The present report is aimed at the registration of the equilibrium EW by the analysis of Brillouin light scattering spectra. Two types of EW are examined. In the first the EW has the form of an unusual case of the transverse bulk wave propagating in a given plane of the unbounded crystal satisfying for certain azimuthal directions free boundary conditions. The other one corresponds to the isolated azimuthal direction on the surface for which the LM degenerates into an EW. Both these situations take place in high-symmetry cuts of cubic crystals having elastic anisotropy ratio $\mu = 2C_{44}/(C_{11} - C_{12}) > 1$ [15], where C_{ij} is an elastic modulus. The shear horizontal bulk mode, propagating along azimuthal directions parallel to the (110) plane, has displacements always in the plane, and confines free boundary conditions along the $[1\bar{1}0]$ axis representing the first EW [16]. The second type of EW is realized on the (111) plane for the $[1\bar{2}1]$ propagation direction where the LM transforms into a bulk plane wave satisfying free boundary conditions. In contrast with the previous case this EW has a propagation vector inclined up the surface [15]. Both EWs considered have velocities in the intermediate bulk velocity range and do not correspond to the limiting modes [17, 18].

0953-8984/94/183359+10$19.50

2. Selection of the optimum scattering configuration

In order to register EWs by the Brillouin spectroscopy method one should specify the samples to be investigated according to their optical properties. That is, Brillouin spectroscopy successfully detects phonon components confining the surface when the contribution of traditional bulk elasto-optic scattering is small. This is easily realized in an opaque medium where the scattering volume is limited by its depth by a few wavelengths [1, 2].

However, highly opaque materials such as metals demonstrate ripple surface light scattering sensitive only to shear vertical displacements [19, 20]. Shear transverse excitations peculiar to EWs, as well as the longitudinal excitations, significantly contribute surface scattered light spectral content via subsurface elasto-optic coupling [21–23]. These controversial requirements, i.e. weak efficiency of traditional bulk scattering but enough intensive subsurface elasto-optic coupling, are found to be met in GaAs. In these crystals Brillouin satellites connecting with an RM of mainly shear vertical excitations were detected together with the spectral lines corresponding to LR of pure longitudinal origin [1, 3, 8, 9].

A revision of the surface scattering polarization configuration is also necessary. Shear horizontal surface excitations 'become visible' only for mixed, i.e. p – s (s – p) scattering configuration [24]. Here the first index corresponds to the polarization of the incident light beam, and the other to that of the scattered one; p means the beam polarized in the plane of incidence and s that perpendicular to it.

The results of cross section calculations for the (110) plane and $[1\bar{1}0]$ propagation direction for different polarization configurations, i.e. p – p, p – s, and s – s according to [24] are shown in figure 1(*a*)–(*c*), respectively. Cross section data for the alternative GaAs crystal orientation, (111) plane, $[1\bar{2}1]$ propagation direction, are qualitatively similar and are omitted.

It is seen from figure 1 that the traditional p – p + s polarization configuration is ineffective for EW registration, the p – s cross section being negligible in comparison with that corresponding to shear vertical (ripple scattering mechanism) and longitudinal surface displacements (subsurface elasto-optic coupling) of the p – p scattering contribution. As a result, components of light scattered by shear horizontal surface excitations should be strongly damped, being practically unresolved from relatively intense components of light scattered by shear vertical or longitudinal surface excitations. The latter is confirmed by the results of the previous GaAs(110) Brillouin spectroscopy observations conducted for the p – p + s polarization configuration where no EWs were detected [1, 8].

In order to observe the EWs one should choose the s – p + s, or s – p (p – s) scattering configuration, and light incidence angle values $\alpha = 45°$–$60°$ at which the ripple scattering contribution falls down, the longitudinal mode originated elasto-optic contribution being comparable with that of shear horizontal surface displacements (s – p + s scattering, figure 1(*b*), (*c*)), or where no ripple or longitudinal elasto-optic contributions were predicted (s – p (p – s) scattering, figure 1(*b*)).

Table 1. Values of elastic moduli, mass density, and dielectric function parameters of GaAs used in the calculations.

Elastic constants [34] (10^{11} N m^2)			Elasto-optic constants [24]			Dielectric function [24]	Mass density [34] (kg m^3)
C_{11}	C_{12}	C_{44}	k_{11}	k_{12}	k_{44}	ϵ_0	d_0
1.188	0.538	0.585	42.5	51.0	25.5	$\simeq 18$	5230.0

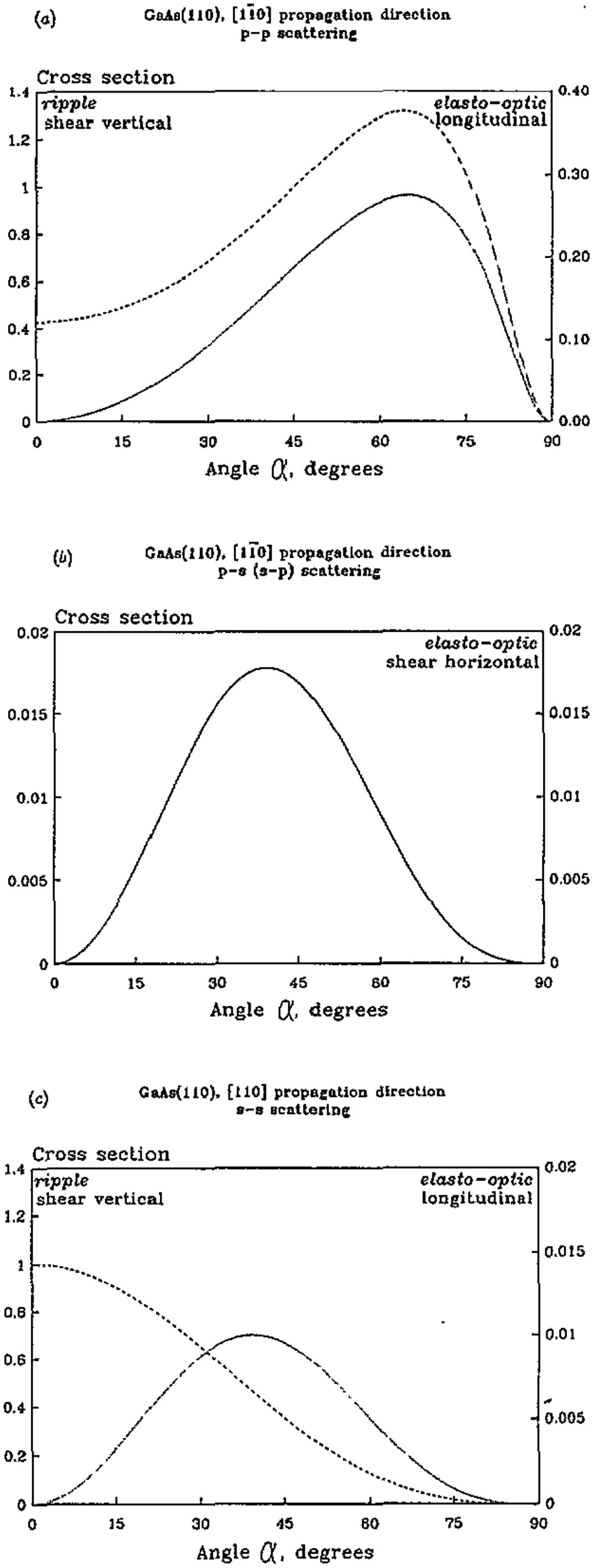
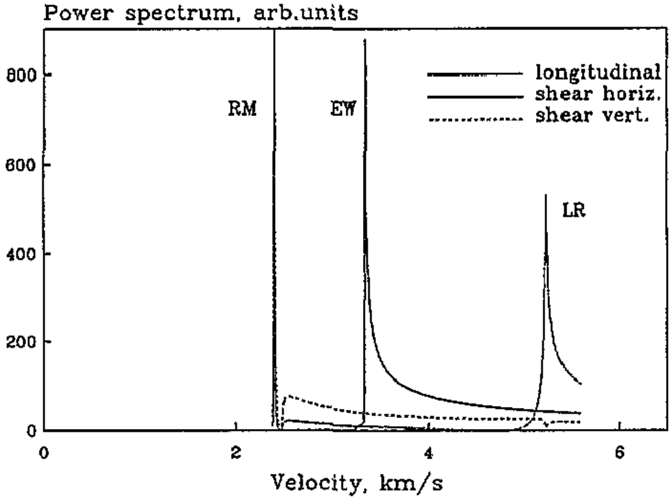


Figure 1. Light scattering cross sections for GaAs crystal ((110) cut, [1$\bar{1}$0] propagation direction) for different types of surface excitation, i.e. longitudinal (elasto-optic coupling; dotted lines), shear horizontal (elasto-optic coupling, solid line), and shear vertical ones (ripple mechanism, dashed line), versus the light incidence angle α. The elasto-optic coupling cross section scale and the type of displacement (longitudinal, or shear horizontal) is given at the right ordinate axis.

(*a*) GaAs(110), $[1\bar{1}0]$ propagation direction



(*b*) GaAs(111), $[1\bar{2}1]$ propagation direction

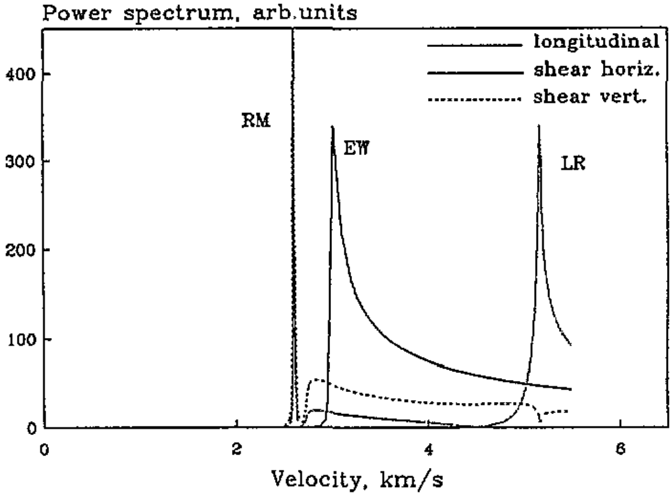


Figure 2. Calculated power spectra of surface excitations of GaAs crystal (*a*) of (110) cut, $[1\bar{1}0]$ propagation direction, and (*b*) of (111) cut, $[1\bar{2}1]$ propagation direction. Three components of the calculated spectra are shown by different line types, see the inset. The dotted line corresponds to longitudinal surface excitation projection, solid, to shear horizontal, and dashed, to shear vertical ones. Material parameters used in the calculations are represented in table 1. Density peaks corresponding to Rayleigh mode, exceptional wave, and longitudinal resonance are marked as RM, EW, and LR, respectively.

Power spectra of surface displacement components for two GaAs crystal orientations, (110) plane, $[1\bar{1}0]$ propagation direction, and (111) plane, $[1\bar{2}1]$ propagation direction, were calculated using surface Green function matching formalism [25, 26], see figure 2(*a*), (*b*).

Different line types correspond to longitudinal, shear horizontal, and shear vertical components of surface displacements respectively, see the figure. Both power spectra reveal strong shear horizontal density peaks corresponding to EWs and having the same intensity scale as that of the longitudinal components of LR already being registered.

3. Incremental technique

The absolute values of cross sections for scattering from shear horizontal surface excitations are very small as compared to the traditional p − p scattering configuration (figure 1(*a*), (*b*)), so high stability of the registration device is required during the light scattering spectra accumulation period. We used the five-pass piezoscanned Fabry–Perot interferometer of Burleigh in our research scheme. Its capability of registering weak Brillouin lines of surface origin in experiments with layered structures was demonstrated earlier [27, 28] at a certain film thickness destructive interference of light scattered by both layer surfaces took place [29, 30]. A backscattering geometry was employed. The scattering was excited by 50–100 mW of linearly polarized light from a single-frequency argon ion laser beam of the $l = 514.5$ nm line. The majority of the experiments was conducted with an interferometer free spectral range, FSR $\simeq 38.0$ GHz. Polarization of the incident light beam as well as the scattered light polarization were specified according to the experiment. See also [31] for details.

Specially prepared low-dislocation specimens of GaAs chosen for the investigations had highly smooth faces parallel to (110) or (111) crystallographic planes. Misorientation of the working faces of the crystals was controlled by x-ray measurements and was less than 1°–2°.

4. Results and discussion

Figure 3 represent examples of the reference Brillouin spectra for the p − p + s scattering configuration registered with the GaAs crystal: (*a*) (110) plane, $[1\bar{1}0]$ propagation direction and (*b*) (111) plane, $[1\bar{2}1]$ propagation direction. Here Brillouin frequency shifts δf are also given in velocity units, V, $V = \delta f l/(2\sin(\alpha))$. The presence of the Brillouin satellite pedestals in the figures is due to light scattering data accumulation conditions. Theoretical cross sections shown with solid lines were calculated according to [24] using the surface phonon power spectra of figure 2 and the instrumental function of the interferometer [32].

It is seen from figure 3 that both (*a*) and (*b*) spectra contain principal Brillouin satellites corresponding to the RM accompanied by relatively weak lines of LR origin.

No Brillouin satellites associated with EW density peaks at $V = 3.35$ km s^{-1} (figure 2(*a*)), and at $V = 3.02$ km s^{-1} (figure 2(*b*)) were predicted or detected experimentally, good agreement between experimental data and theoretical cross section being achieved for the entire frequency region. The asymmetry and high-frequency broadening of the principal satellites are mainly explained by the presence of relatively intense bulk threshold shear vertical phonon components at $V \geqslant 2.48$ km s^{-1} (figure 2(*a*)), and at $V \geqslant 2.73$ km s^{-1} (figure 2(*b*)).

Brillouin spectra of light scattered from the (110) face of a GaAs crystal, $[1\bar{1}0]$ propagation direction, for s − p + s and s − p polarization configurations are shown in figure 4(*a*), (*b*), respectively.

Here in both spectra Brillouin satellites corresponding to EW appear, coupled in figure 4(*a*) with the lines of RM and LR origin, and being a single component of the registered

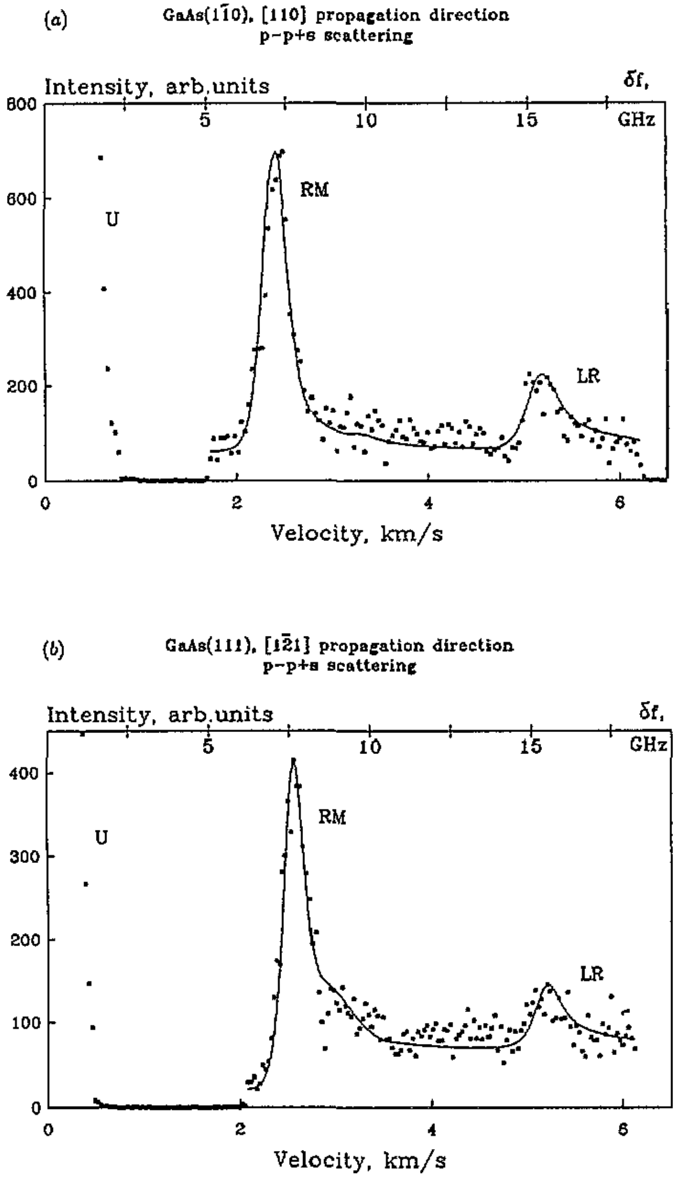


Figure 3. Brillouin spectrum of light scattered by GaAs crystal (*a*) of (110) and (*b*) of (111) cut. The surface acoustic wave vector is parallel to [1$\bar{1}$0] and [1$\bar{2}$1] respectively. $\alpha = 50°$, $l = 514.5$ nm, $p - p + s$ scattering polarization configuration. Brillouin lines corresponding to Rayleigh wave, leaky mode and longitudinal resonance are marked as RM, LM, and LR. U is an unshifted line. The theoretical cross section calculated using data of table 1 is shown by a solid line.

spectrum of figure 4(*b*). The theoretical cross section reasonably fits the experimental data for both spectra represented. Additionally, the EW satellite frequency shift follows with experimental accuracy a $\sin(\alpha)$ law specific to the surface Brillouin scattering. Both these facts: the coincidence of Brillouin experimental data spectral content with those of

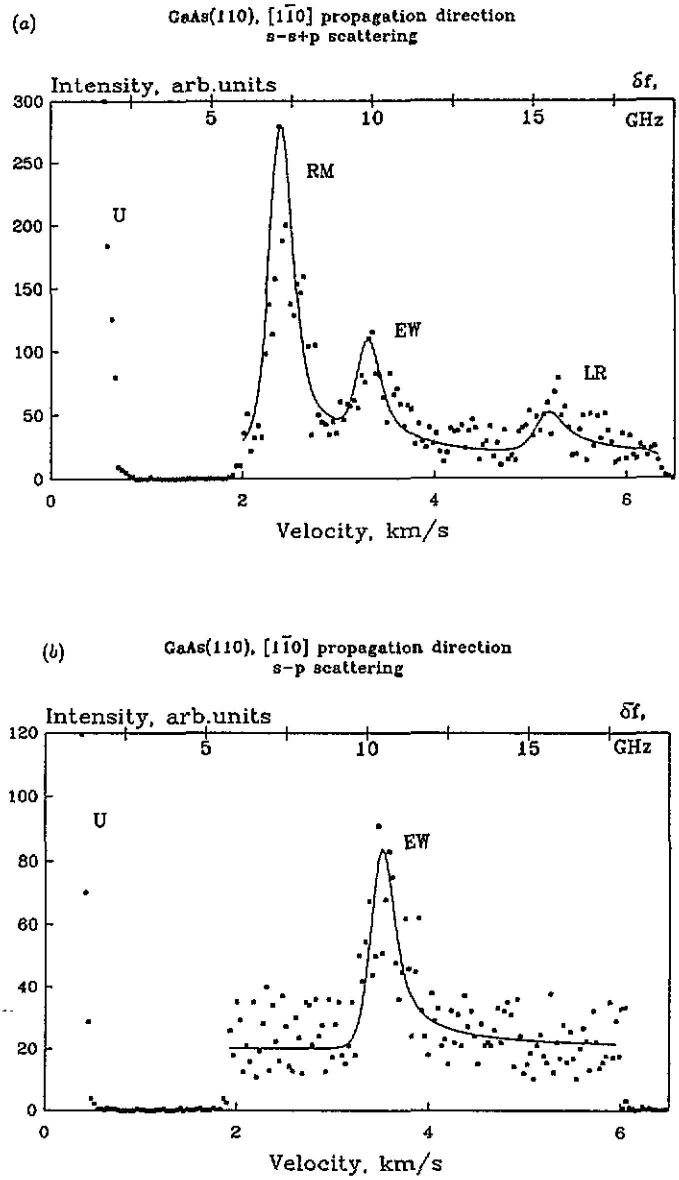
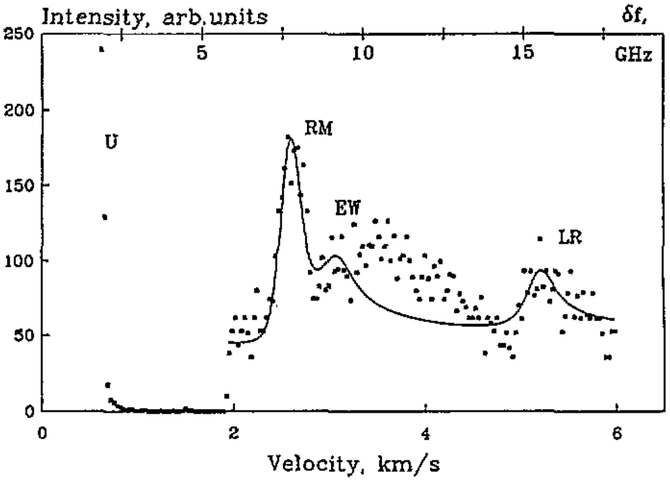


Figure 4. Brillouin spectrum of light scattered by GaAs crystal of (110) cut. The surface acoustic wave vector is parallel to [1$\bar{1}$0]. (*a*) s − p + s, (*b*) s − p scattering polarization configuration. The Brillouin line corresponding to the exceptional wave is marked as EW. Same conventions as in figure 3.

theoretical cross section, and the variation of the satellite frequency shift as sin(α), confirm that this line is of unique EW origin. The discrepancy between the calculated EW velocity value and the position of the maxima of the Brillouin peaks does not exceed 2%.

Three Brillouin satellites were registered in the spectrum of figure 5(*a*) (GaAs(111), [1$\bar{2}$1] propagation direction, s − s + p polarization configuration) similarly to those of figure 4(*a*). However, the spectral line of scattered light of EW velocity range

(*a*) GaAs(111), [$1\bar{2}1$] propagation direction
s-p+s scattering



(*b*) GaAs(111), [$1\bar{2}1$] propagation direction
s-p scattering

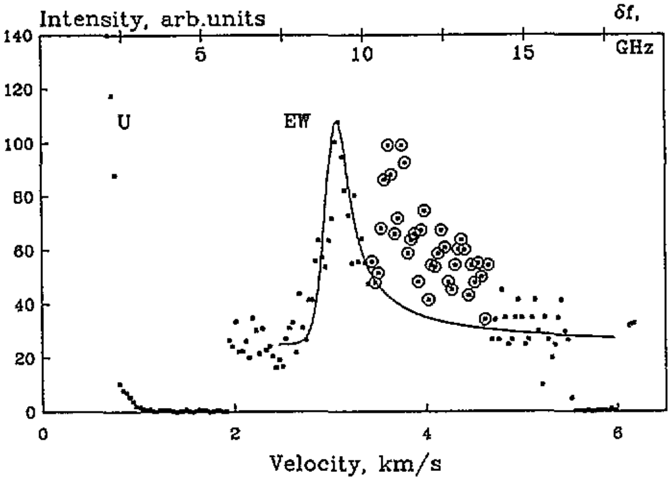


Figure 5. Brillouin spectrum of light scattered by GaAs crystal of (111) cut. The surface acoustic wave vector is parallel to [$1\bar{2}1$]. (*a*) s − p + s, (*b*) s − p scattering polarization configuration. Same conventions as in figure 3.

$V = 3.0$–4.0 km s^{-1} appears to be relatively wide, and the position of its maximum has a velocity remarkably higher than that of the EW for all values of α. The corresponding theoretical cross section shows a mismatch with experimental data for this velocity range.

Registration of the spectra with s − p polarization configuration, figure 5(*b*), when scattering at RM and LM is forbidden [24] and so the $V = 3.0$–4.0 km s^{-1} velocity region is not perturbed by the tails of corresponding Brillouin satellites, reveals that there exist two spectral lines instead of one. The first one corresponds to the EW, the theoretical curve for its cross section satisfactorily coinciding with the experimental points. The frequency shift of this Brillouin satellite follows the sin(α) law in the whole α region, $\alpha = 45°–60°$, which confirms its surface scattering nature, and consequently the EW origination. The position of the Brillouin peak corresponding to the EW coincides with the calculated EW velocity value within 1–3%.

Special care has been taken to trace the source of the second component of the light scattering spectrum marked by circles. The position of this extra line does not vary with change of α. We have proved by the interferometer FSR variation that this line has 'true' $\delta f \simeq 49.0$ GHz. It corresponds to the transverse bulk wave propagating along the direction close to $[1\bar{1}1]$ which satisfies simple 'bulk mode' Brillouin spectroscopy conditions [33]. The position of this line in the Brillouin spectrum of figure 5(*b*) belongs to the neighbouring interferometer order.

Finally, observation of light scattered from the (110) and (111) planes of a GaAs crystal for $\mathrm{s} - \mathrm{s} + \mathrm{p}$ and $\mathrm{s} - \mathrm{p}$ polarization configuration reveals a new type of Brillouin line of surface origin. These new lines are attributed to the EWs propagating along $[1\bar{1}0]$ and $[1\bar{2}1]$ crystallographic directions, respectively. The results of theoretical calculations describe scattered light spectral content reasonably well.

Acknowledgments

The authors are very grateful to Dr V M Avdiuhina for perfect GaAs samples, M Saphonov for his help during the experiments, Professors V R Velasco, F Nizzoli, A M Diakonov, V I Alshits, and Dr A L Shuvalov for fruitful discussions, and to Professor I A Yakovlev and Dr T S Velichkina for their interest in our work. VVA is also thankful to Profesors V R Velasco and F Garcia-Moliner for their hospitality during his stay in Madrid, October 20 to November 6, 1993. The contribution of VVA was partially supported by NATO under Collaborative Research Grant No 930164.

References

[1] Sandercock J R 1978 *Solid State Commun.* **26** 547

[2] Nizzoli F and Sandercock J R 1990 *Dynamical Properties of Solids* vol 6 ed H K Horton and A A Maradudin (Amsterdam: North-Holland) p 281

[3] Carlotti G, Fioretto D, Giovannini D, Nizzoli F, Socino G and Verdini L 1992 *J. Phys.: Condens. Matter* **4** 257

[4] Baumgart P, Blumenroder S, Erle A, Hillebrands B, Guntherodt G and Schmidt H 1989 *Solid State Commun.* **69** 1135

[5] Boekholt M, Harzer J V, Hillebrands B and Guntherodt G 1991 *Physica* C **179** 101

[6] Aleksandrov V V, Velichkina T S, Mozhaev V G and Yakovlev I A 1991 *Solid State Commun.* **77** 559

[7] Aleksandrov V V, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Phys. Lett.* **171A** 103

[8] Aleksandrov V V, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Phys. Lett.* **170A** 165

[9] Aleksandrov V V, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Zh. Eksp. Teor. Fiz.* **102** 1891 (Engl. Transl. 1992 *Sov. Phys.–JETP* **102** 1019)

[10] Aleksandrov V V, Velichkina T S, Vorob'ev P G Potapova Ju B and Yakovlev I A 1993 *Zh Eksp. Teor. Fiz.* **103** 2170 (Engl. Transl. 1993 *Sov. Phys.–JETP* **76** 1085)

[11] Aleksandrov V V, Gladkevitch A V, Mozhaev V G, Giovannini L and Nizzoli F to be published

[12] Chadwick P and Smith G P 1977 *Adv. Appl. Mech.* **17** 303

[13] Barnett D M and Lothe J 1985 *Phys. Rev. Lett.* A **402** 135

[14] Chadwick P 1985 *Phys. Rev. Lett.* A **401** 203

[15] Farnell G W 1970 *Physical Acoustics* vol VI ed W P Mason and R N Thurston (New York: Academic) p 109

[16] The other EW propagating along [001] at the (110) surface [15] is not considered in the current research.

[17] Barnett D M, Lothe J, Nishioke K, Asaro R J, 1973 *J. Phys. F: Met. Phys.* 3 1083

[18] Alshits V I, Darinskii A N and Shuvalov A L 1991 *Statistical Physics and Dislocation Theory* ed T Jossang and D M Barnett (Oslo: Oslo University Press) p 189

[19] Loudon R 1978 *PRL* **40** 581

[20] Loudon R and Sandercock J R 1980 *J. Phys. C: Solid State Phys.* **13** 2609

[21] Bortolani V, Nizzoli F and Santoro G 1978 *Phys. Rev. Lett.* **41** 39

[22] Camley R E and Nizzoli F 1985 *J. Phys. C: Solid State Phys.* **18** 4795

[23] Bassoli L, Nizzoli F and Sandercock J R 1986 *Phys. Rev.* B **34** 1296

[24] Marvin A M, Bortolani V, Nizzoli F and Santoro G 1980 *J. Phys. C: Solid State Phys.* **13** 1607

[25] Velasco V R and Garcia-Moliner F 1980 *J. Phys. C: Solid State Phys.* **13** 2237; 1980 *Solid State Commun.* **33** 1

[26] Garcia-Moliner F and Velasco V R 1992 *Theory of single and multiple interfaces* (Singapore: World Scientific)

[27] Aleksandrov V V, Potapova Yu B, Vorob'ev P A, Dyakonov A M and Yakovlev N L 1993 *Fiz. Tverd. Tela* **35** 2437 (Engl. Transl. 1993 *Sov. Phys.–Solid State* **35** 1206

[28] Aleksandrov V V, Saphonov M V, Velasco V R, Yakovlev N L and Martynenko L Ph to be published

[29] Bortolani V, Marvin A M, Nizzoli F and Santoro G 1983 *J. Phys. C: Solid State Phys.* **16** 1757

[30] Karanikas J M, Sooryakumar R and Phillips J M 1989 *Phys. Rev.* B **39** 1388

[31] Aleksandrov V V, Velichkina T S, Voronkova V I, Diakonov A M, Symikov P P, Yakovlev I A and Yanovskii V K 1989 *Phys. Lett.* A **142** 307

[32] Lindsay S M, Burgers S and Shepherd I W 1977 *Appl. Opt.* **16** 1404

[33] Fabelinsky I L 1968 *Molecular Scattering of Light* (New York: Plenum)

[34] Slobodnik A J Jr, Conway E D and Delmonico R T 1973 *Surface Wave Velocities (Microwave Acoustics Handbook) 1A (Hanscom AFB, MA: Air Force Cambridge Research Laboratories)*