

# The 45th anniversary of the Institute of Spectroscopy of the Russian Academy of Sciences (Scientific session of the Physical Sciences Division of the Russian Academy of Sciences, 23 October 2013)

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On 23 October 2013, the out-of-town scientific session of the Physical Sciences Division of the Russian Academy of Sciences (RAS), celebrating the 45th anniversary of the Institute of Spectroscopy, RAS (ISAN in *Russ. abbr.*), was held at the conference hall of the Institute's building in Troitsk.

The agenda of the session, announced on the website [www.gpad.ac.ru](http://www.gpad.ac.ru) of the Physical Sciences Division, RAS, listed the following reports:

(1) **Vinogradov E A** (ISAN, Troitsk, Moscow) “The main achievements of ISAN from 2009 to 2013”;

(2) **Balykin V I** (ISAN, Troitsk, Moscow) “Atomic and photonic nanooptics”;

(3) **Lozovik Yu E** (ISAN, Troitsk, Moscow) “Electronic and collective properties of topological isolators”;

(4) **Yevdokimov Yu M** (Engelgard Institute of Molecular Biology, RAS, Moscow), **Kompanets O N** (ISAN, Troitsk, Moscow) “Structural nanotechnology of DNA (liquid-crystal approach) and its applications”;

(5) **Chekalin S V** (ISAN, Troitsk, Moscow) “Femtosecond spectroscopy of promising materials”.

Papers written on the basis of oral reports 1, 2, 4, and 5 are published below.

PACS numbers: **05.40.**–a, 42.50.Lc, **71.36.**+c  
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## Near field phonon–polariton spectroscopy as a method for studying the optical properties of nanofilms

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### 1. Introduction

The use of thin wide-gap semiconductor films for various applications, in particular, in optoelectronics, requires knowledge of their physical properties. However, traditional reflection, transmission, and reflection–absorption spectra (far-field spectra) of structures with thin films cannot give the required information. The photometric accuracy of

modern spectrometers is still not sufficient for studying the physical properties of such samples, because a weak absorption spectrum of a thin film is masked by the background spectrum of a substrate. In a number of cases, the physical parameters of nanofilms can be determined by applying surface polariton spectroscopy. Surface polaritons (SPs) are nonradiative electromagnetic excitations propagating along the medium interface, with the SP field concentrating directly near the interface and exponentially decaying with distance from the interface [1–4].

The SP field near the film surface (interface) is a near field with the strength considerably exceeding that of the fluctuation thermal far field [1–3]. Because of this property, the SP spectra can be very sensitive to the characteristics of the interface. This is especially important, because the properties of films in most cases greatly differ from those of the bulk materials from which they are fabricated and, additionally, the structure of the near-surface layer of a substrate can change during the deposition (growth) of films [5].

The analytical possibilities of the SP spectroscopy drastically increase if the frequencies of optical phonons of the film fall within the frequency range of SPs on the substrate. The frequency resonance causes splitting and a shift of the SP spectra in the substrate. In this case, experimental data allow one to determine the permittivities of both the film and substrate, the film thickness, and to specify its structure. A gap appearing in the dispersion curves of SPs in the substrate is proportional to the square root of the film thickness, thus allowing the estimation of the thickness of very thin films [4, 6].

In this paper, we discuss the spectroscopic studies of surface phonon polaritons as a unique source of information about the physical properties of ultrathin films on the surfaces of single crystals.

### 2. Experimental results

As an example, we consider the experimental results of studying the optical properties of MgO films (10, 30, 100, and 300 nm in thickness) [7] and AlN films (2, 40, and 400 nm

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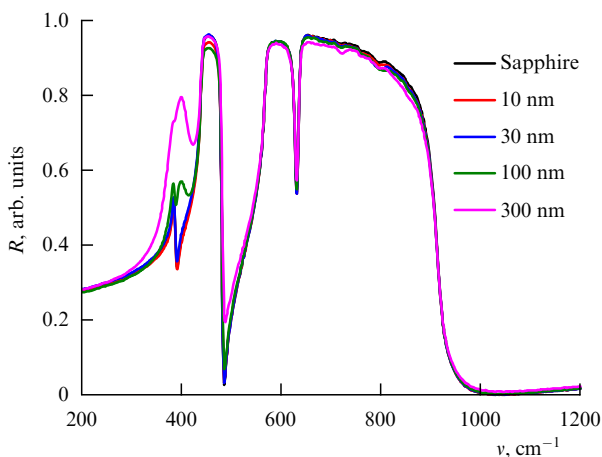
in thickness) [8, 9] on sapphire substrates (the optical axis of the substrate is perpendicular to its surface). External reflection spectra (in the far field) were measured with a Bruker IFS66v IR Fourier spectrometer at angles close to the normal incidence.

Surface polaritons—near-field electromagnetic excitations—belong to nonradiative excitations with the wave vector exceeding the wave vector of light in a vacuum; therefore, SPs do not interact with the external electromagnetic field under normal conditions [10, 11]. An ATR (attenuated total reflection) prism mounted with some gap above a sample makes possible the matching between the wave vectors of an SP and exciting light, thereby ensuring the resonance absorption of light by the SP. In this case, the SP field is disturbed by the prism, and the SP electromagnetic field, making up the near field in the absence of the prism, is transformed by the prism to the far field which is recorded with a standard IR spectrometer [1, 2, 4]. The SP spectra were taken using a thallium halide single-crystal (KRS-5) ATR prism mounted over a film on a sapphire substrate with different gaps and an IFS66v IR Fourier spectrometer at different angles (larger than the critical angle) of incidence of light in the prism [1, 2, 4].

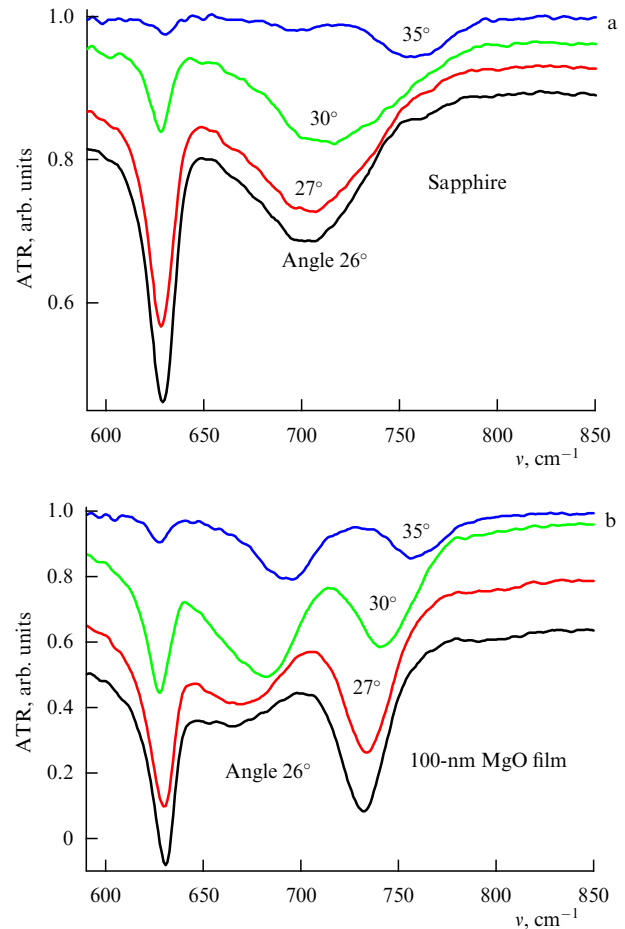
MgO layers 10 and 30 nm in thickness are not seen in the far-field reflection spectra of MgO films on sapphire (Fig. 1). For film thicknesses of 100 and 300 nm, the reflection spectrum changes only in the low-frequency region (near the frequency of  $400\text{ cm}^{-1}$ ) due to the resonance of the transverse optical (TO) phonon of magnesium oxide and a low-frequency TO phonon of sapphire. These experimental data are not sufficient even for comparatively thick MgO films for correct measurements of the film permittivity.

Figure 2 shows near-field ATR spectra for different angles of light incidence inside a prism for initial sapphire and sapphire with a 100-nm-thick MgO film. Note that the optimal gap between the ATR prism and a sample surface should be on the order of the SP wavelength, which is inversely proportional to the SP wave vector parallel to the interface. The SP wave vector is, in turn, proportional to both the refractive index of the prism material and the sine of the angle of incidence of light in the prism.

Absorption bands in the ATR spectra of sapphire are caused by SPs at the sapphire–vacuum interface. The



**Figure 1.** (Color online.) Far-field reflection spectra of 10-, 30-, 100-, and 300-nm-thick MgO films on sapphire for normally incident light [7].



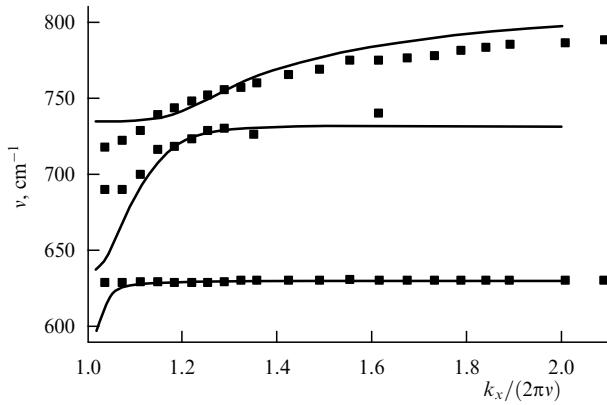
**Figure 2.** ATR spectra of (a) sapphire and (b) a 100-nm-thick MgO film on sapphire for four angles of incidence of light in a KRS-5 prism (spectra are shifted along the ordinate axis to avoid their overlapping).

dependences of the frequencies of these bands on the angle of incidence of light in the prism give the SP dispersion, i.e., the wave-vector dependence of the frequency [7–11].

Figure 2b demonstrates that the MgO film drastically changes the SP spectrum of sapphire due to the occurrence of resonance between the SP frequency in sapphire and a longitudinal optical phonon in the MgO film. This interaction causes the splitting of the high-frequency branch of the SP spectrum in sapphire at frequencies near  $700\text{ cm}^{-1}$ .

The ATR spectra are known to be very sensitive to a gap between the sample and prism, because the latter disturbs the SP field. To measure correctly the frequency and half-width of an SP absorption band for different angles of incidence of light in the prism (different SP wave vectors), it is necessary to select experimentally the gap that provides the intensity of the band being measured of about a few percent. In this case, the perturbation of the SP by the prism is weak.

The frequency-angular dependence of the SP determines its dispersion. The SP dispersion is well described theoretically if the permittivities of the film and substrate and the film thickness are known. The dispersion of SPs in sapphire with film was recovered from the angular dependence of ATR spectra. This dispersion was compared with the SP dispersion calculated in Ref. [4] using the sapphire permittivity [obtained from the dispersion analysis of the external reflection spectra of sapphire without film (see Fig. 1 and Fig. 4 below)], whose constants coincide within the limits of experimental error with



**Figure 3.** Dispersion of surface polaritons in a 10-nm-thick MgO film on sapphire. Full squares are experimental data, and solid curves are calculated using optical constants of bulk MgO and sapphire.

those in Ref. [12]. The parameters of the permittivity of the film in these calculations were varied to obtain the best fit between the calculated SP dispersion and experimental frequency–angular spectra. Figure 3 depicts the dispersion curves of SPs in a 10-nm-thick MgO film on sapphire, which could not be observed in far-field spectra.

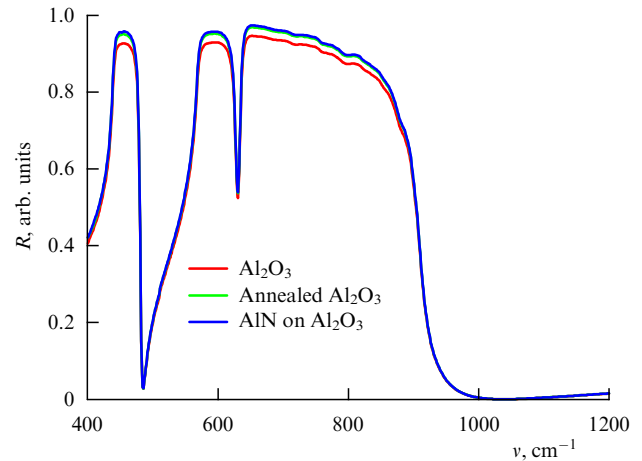
### 3. Discussion

The dispersion curves of SPs, similar to those presented in Fig. 3, were also obtained for 30-, 100-, and 300-nm-thick MgO films [7]. The best agreement between theoretical dispersion curves, achieved by varying in calculations the frequencies of optical phonons in films, their oscillator strengths, and decay constants, allowed us to recover the optical constants (permittivities) of films on sapphire.

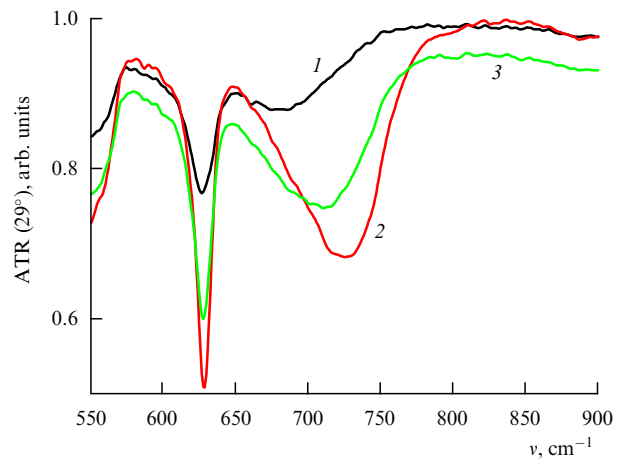
It should be noted that the optical constants of 10-nm-thick films proved to be very close to those for bulk MgO, unlike the optical constants of thicker films. It is well known that good-quality films grow without strong stresses if the lattice constants of the film and substrate are close to each other. Otherwise, strong stresses emerge during the film growth, which shift the frequencies of optical phonons, changing their oscillator strengths and decay constants. In addition, these stresses give rise to so-called misfit dislocations, reducing stresses in films but deteriorating some properties of the film material important for applications.

Notice also that correct measurements of the spectra at many angles of incidence of light inside a prism make, in fact, the ATR spectroscopy a quantitative technique for measuring the permittivity of ultrathin films, also including monolayer films for which the concept of the macroscopic permittivity seems to be inapplicable. Nevertheless, as is seen from the results presented above, the permittivity of the 10-nm-thick MgO film is well described by the standard model of the permittivity of a bulk MgO.

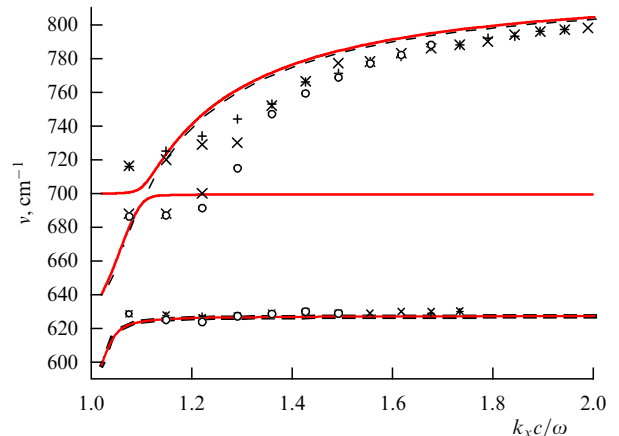
An interesting result was also obtained for monolayer AlN films on sapphire [8]. The annealing of sapphire crystals in a nitrogen flow at temperatures above 900 °C leads to the replacement of oxygen in sapphire by nitrogen and the growth of AlN monolayers layer-by-layer [8, 9]. Figure 4 shows the external reflection spectra of sapphire before and after its annealing and during nitridization. These spectra almost coincide.



**Figure 4.** External reflection spectra of sapphire single crystals before and after annealing, and after sapphire nitridization [8].



**Figure 5.** ATR spectra for unannealed (curve 1), annealed (curve 2), and nitridized (curve 3) sapphire.



**Figure 6.** (Color online.) Dispersion of surface polaritons of unannealed (circles), annealed (crosses), and nitridized (x's) sapphire. Curves are calculated for sapphire without a film (black dashed curves) and sapphire with a 2-nm-thick AlN film (solid red curves).

In a ‘near-field’ part of ATR spectra (Fig. 5) taken around 700 cm<sup>−1</sup> (close to the frequency of the transverse optical phonon in AlN), as in the case of MgO films on sapphire (Fig. 2b), the splitting of the dispersion curve of SPs in sapphire was observed [8, 9] (Fig. 6). From these experi-

mental data, the frequencies of transverse phonons ( $700\text{ cm}^{-1}$ ) and longitudinal oscillations ( $890\text{ cm}^{-1}$ ), as well as their oscillator strengths, were obtained for 1- and 2-nm-thick AlN films [8] and for 25- and 400-nm-thick AlN films on sapphire [9].

It would seem that follow-up studies of the optical characteristics of AlN during monolayer-by-monolayer growth would provide answers to very important questions, namely, how the AlN layers are formed, how their characteristics change due to great stresses in them caused by a large mismatch between lattice constants, and how the surface layers of sapphire are transformed.

Unfortunately, direct surface-nonperturbing methods for measuring the fluctuating fields of surface electromagnetic excitations are absent at present, and the near-field spectroscopy (in the ATR regime) appears to be the most ‘delicate’ technique for studying the surface of crystals and thin films deposited on them.

#### 4. Conclusion

Correct measurements of ATR spectra for many angles of incidence of light inside a prism make ATR spectroscopy, in fact, a quantitative technique for measuring the permittivity of ultrathin films, including monolayer films, in which the concept of the macroscopic permittivity appears inapplicable. Nevertheless, as is seen from the results presented above, 10-nm-thick MgO film and 1- and 2-nm-thick AlN films are well described by the standard model of the permittivity of bulk MgO and AlN, respectively.

In this paper, we have studied the resonance interaction of the substrate SPs with optical phonons in films and measured the splitting of SP dispersion curves. This splitting was proved to be proportional to the square root of the film thickness. It is shown that the ATR spectroscopy can be utilized for determining the permittivity of thin and ultrathin films.

#### Acknowledgments

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## Quantum control of atoms and photons by optical nanofibers

V I Balykin

### 1. Introduction

Studies of the interaction of light with matter at the level of single atoms and photons not only provide a deeper understanding of the quantum-mechanical nature of the light–matter interaction but also open up prospects for new quantum technologies. During the last two decades, quite a few approaches have been adopted for realizing interaction at the level of single atoms and photons. The possibility of achieving single-particle interaction is basic to the concept of quantum information processing and communication [1, 2]: atoms are treated as physical objects that can store and process information, and photons as objects for long-range data transmission through a quantum information network. The quantum information network can comprise, for instance, single atoms (ions, molecules, or quantum dots) representing addressable points of the quantum information network and optical (or other) waveguides providing photon-assisted effective connections among nodes of the quantum information network.

At present, the efficient coupling of single photons with an atomic medium is achieved on a basis of the following physical mechanisms: (1) the microscopic localization of single atoms and the attainment of the ‘strong interaction’ regime [3, 4]; (2) the use of laser fields focused to light spots comparable in size to the absorption cross section of a single atom [5–8], and (3) the employment of coherent coupling with optically thick atomic ensembles [2]. Despite the outstanding achievements obtained by these methods, a simple, reliable, and scalable technique for the integration of simple quantum systems with photon systems has been lacking.

In this paper, we will briefly review the work on the deterministic control of single atoms and single photons, and on establishing conditions for their efficient interaction. Such a control based on the use of *ultracold atomic ensembles near an optical nanowaveguide* was first proposed a decade ago by Balykin et al. [9]. The subsequent experimental realizations of a hybrid (atom + nanowaveguide) system [10–12] have demonstrated the outlook for its use in a variety of applica-

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