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Nanoplasmonics and metamaterials
(Scientific session of the Division of Physical Sciences, Russian Academy of Sciences, 27 April 2009)

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On 27 April 2009, in the conference hall of the Lebedev Physical Institute, Russian Academy of Sciences, a scientific session of the Division of Physical Sciences of the Russian Academy of Sciences devoted to the problem of nanoplasmonics and metamaterials took place. The following reports were presented at the session:

(1) **Tikhodeev S G, Gippius N A** (Prokhorov Institute of General Physics, Russian Academy of Sciences, Moscow) "Plasmon–polariton effects in nanostructured metal–dielectric photonic crystals and metamaterials";

(2) **Shubina T V, Ivanov S V, Toropov A A, Kop'ev P S** (Ioffe Physicotechnical Institute, Russian Academy of Sciences, St. Petersburg) "Plasmon effects in In(Ga)N-based nanostructures";

(3) **Kurin V V** (Institute of Physics of Microstructures, Russian Academy of Sciences, Nizhnii Novgorod) "Resonance scattering of light in nanostructured metallic and ferromagnetic films";

(4) **Lagarkov A N , Sarychev A K** (Institute of Theoretical and Applied Electrodynamics, Joint Institute of High Temperatures, Russian Academy of Sciences, Moscow) "Active optical metamaterials";

(5) **Gippius N A, Tikhodeev S G** (Prokhorov Institute of General Physics, Russian Academy of Sciences, Moscow) "Application of the scattering matrix method for calculating the optical properties of metamaterials."

Summaries of reports 1–3 and 5 and of an article written on the basis of report 4 are given below.

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Plasmon–polariton effects in nanostructured metal–dielectric photonic crystals and metamaterials

S G Tikhodeev, N A Gippius

In this report, we give a brief introduction to nanoplasmonics, the optical properties of plasmon–polariton photonic crystals, and metamaterials. In more detail, we analyze the problem of determining the effective electromagnetic response of thin layers of metamaterials of an arbitrary symmetry, including gyrotropic ones.

Surface plasmons are collective oscillations of conduction electrons of a metal excited by light near its surface — the interface with a dielectric. Depending on the geometry of the metal–dielectric structure, localized plasmons (in metal clusters or pores inside metal with a size much smaller than the wavelength of light, i.e., in nanoclusters) and delocalized surface plasmons (on an infinite flat metal–dielectric interface) are distinguished. However, even in the case of delocalized plasmons, their excitation by light requires that the metal surface be nanostructured. Therefore, this thriving field of physics is frequently called *nanoplasmonics*.

In fact, nanoplasmonics has very long been used by humankind. The localized plasmons in silver and gold nanoclusters in glass ensure the extraordinary brightness and longevity of the colored stained-glass windows of medieval cathedrals. We can remember an even older example — the famous Lycurgus Cup, which was made, apparently, in Rome in the 4th century A.D. (now in the British museum); it is also made from glass with metallic nanoclusters. However, the truly thriving development and application of nanoplasmonics started only recently in connection with the development of nanotechnology and computational electrodynamic methods [1–5].

Nanoplasmonics is attractive, first of all, because plasmons allow concentrating electromagnetic energy in small volumes (in comparison with the wavelength of light). Plasmons, having a giant dipole moment, are efficient mediators in the interaction of these small volumes with light. Furthermore, the properties of plasmons can be controlled in extremely wide limits.

A detailed survey of the achievements of nanoplasmonics is beyond the scope of this report. We only mention here that one of the basic methods of controlling plasmons is constructing so-called polaritonic crystals. Polaritonic crystals are artificial periodic media, in which optically active electronic resonances exist together with photonic resonances (which arise due to a periodic modulation of a dielectric constant). The coupled photonic and electronic resonances are conventionally called polaritons; therefore, photonic crystals with interacting electronic and photonic resonances are now called polaritonic crystals. Initially, photonic crystals made of transparent dielectrics with light-frequency-independent dielectric constants were studied [6–9]. In the first polaritonic crystals, Bragg superlattices of

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semiconductor quantum wells were used [10, 11]. The role of electronic resonances was then played by excitons in the quantum wells. Later, exciton–polariton crystals with another geometry were proposed, in the form of so-called photonic-crystal slabs [12–14], i.e., planar waveguide layers modulated by one-dimensional or two-dimensional lattices, e.g., of depressions filled with a layered semiconductor with strong excitonic resonances.

But polaritonic effects in modulated metal–dielectric structures proved to be most interesting. The role of electronic resonance is there played by localized or surface plasmons. The first samples of such 'polaritonic crystalline layers' were investigated more than a hundred years ago; however, they had a different name then, diffraction gratings. The first plasmon–polariton effects were the resonance anomalies found by Wood [15] in the optical spectra of lattices created on the surface of a metal, and first explained by the excitation of surface plasmons by Fano [16].

Subsequently, significant attention has been given to these structures due to the detection of so-called anomalous light transmission through a lattice of subwavelength holes in a metal layer [17]. We also note the formation of plasmon–waveguide polaritons in lattices of metallic nanoclusters or nanowires on the surface of a planar dielectric waveguide [18, 19], as well as interesting plasmonic effects in metal layers with lattices of voids [20, 21].

It was recently revealed that when using ferromagnetic materials (for preparing either a dielectric waveguide or nanoclusters), extremely interesting magnetooptical effects potentially important in applications [22, 23] appear in such systems.

But the greatest burst of interest in metal–dielectric polaritonic crystals arose in connection with the possibility of designing artificial media, so-called metamaterials with a controlled electromagnetic response, on their basis. Among the possibilities discussed are metamaterials with a negative refractive index [24] for creating unconventional new optic devices and new methods of controlling light [3, 25–31].

A medium with a negative refractive index must have negative dielectric and negative magnetic constants (a more precise formulation for absorbing media: different signs of the real and imaginary parts of the refractive index.) The negative sign of the dielectric constant is ensured by the use of a metal. To ensure a negative magnetic susceptibility, structurization is required; it is necessary to ensure a magnetoinductive resonance, which requires the presence of circular current contours. A medium with a negative refractive index was first realized for the microwave range with the aid of split ring resonators [26]. Then it was understood [27] that in the near-infrared and visible optical ranges, the role of ring contours for the current can be played by coupled localized plasmons, for example, based on double-chain metallic nanowires. Metamaterials have been proposed based on pairs of periodically perforated metallic layers, so-called fishnet structures [28, 32], and media with a strong natural optical activity [31, 33, 34] and with strong optical nonlinearities [35, 36] have been created.

Metamaterials are short-period metal–dielectric plasmonic crystals. The idea consists in the period of the metamaterial being less than the relevant wavelength of light. Then the layer of the metamaterial in the far wave zone behaves as a layer of a uniform substance; there is no diffraction, only transmission, reflection, and absorption of light. The question of correctly describing the effective electromagnetic response of metamaterials is therefore very important [37, 38]. One of the methods to obtain the effective dielectric and magnetic susceptibilities (ε and μ) of a metamaterial is the parameterization of the experimentally measured or theoretically calculated coefficients of transmission and reflection of a finite-thickness layer of a metamaterial [39, 40]. Below, we briefly describe the scheme of this parameterization, which becomes highly nontrivial in the case of a metamaterial of lowered symmetry.

It is known that for a complete description of the electromagnetic properties of a homogeneous medium (including one without an inversion center, i.e., gyrotropic), it is sufficient to introduce the nonlocal dielectric constant tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ and to assume that $\mu = 1$ [37]. Such a description — with the aid of a nonlocal dielectric susceptibility — includes, as a special case, the traditional approach to nongyrotropic media with the use of local $\varepsilon_{ij}(\omega)$ and $\mu_{ij}(\omega) \neq 1$ generally accepted for describing media with a negative refractive index.

We consider the most general case of a gyrotropic metamaterial whose gyrotropy occurs exclusively because of nanostructurization without an inversion center, while the substances composing the metamaterial are nongyrotropic (and there is no stationary magnetic field). Then, in addition to the effective $\varepsilon_{ij}(\omega)$ and $\mu_{ij}(\omega)$ tensors, it is necessary to introduce local susceptibilities, which correspond to odd terms in the expansion of the total nonlocal dielectric susceptibility in the powers of $\mathbf{k}$ (beginning with the linear term). These additional susceptibilities, which are sometimes referred to as the coefficients of chirality $\chi(\omega)$ and bianisotropy $\beta(\omega)$, correspond to a linear coupling of the magnetic induction $\mathbf{B}$ to the electric field $\mathbf{E}$ and of the electric induction $\mathbf{D}$ to the magnetic field $\mathbf{H}$ [41–44].

It is known that a necessary condition for the existence of such susceptibilities is the absence of both the inversion center and time reversibility [45]. In the case of metal–dielectric metamaterials, the first condition is ensured by the asymmetry of the unit cell, and the second condition is always fulfilled because metals are absorbing media.

As an example, we consider the case of a plane electromagnetic wave (propagating along the z axis) incident normally on a layer of a metamaterial in the plane xy. Then, in the most general case of a gyrotropic metamaterial consisting of nongyrotropic components, the effective electromagnetic response can be completely described by introducing *ten* linearly independent coefficients of response in the far field, which relate the tangential components of the fields $(\mathbf{D}_{\|}, \mathbf{B}_{\|})$ to $(\mathbf{E}_{\|}, \mathbf{H}_{\|})$,

$$\begin{pmatrix} D_x \\ D_y \\ B_x \\ B_y \end{pmatrix} = \hat{\eta}_{\|}(\omega, \mathbf{k}_{\|})\big|_{\mathbf{k}_{\|}=0} \begin{pmatrix} E_x \\ E_y \\ H_x \\ H_y \end{pmatrix}, \tag{1}$$

where

$$\hat{\eta}_{\|} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \mathrm{i}\chi_x & \mathrm{i}\beta_x \\ \varepsilon_{xy} & \varepsilon_{yy} & -\mathrm{i}\beta_y & -\mathrm{i}\chi_y \\ -\mathrm{i}\chi_x & \mathrm{i}\beta_y & \mu_{xx} & \mu_{xy} \\ -\mathrm{i}\beta_x & \mathrm{i}\chi_y & \mu_{xy} & \mu_{yy} \end{pmatrix}. \tag{2}$$

The ten components ε_{xx}, ε_{yy}, ε_{xy}, μ_{xx}, μ_{yy}, μ_{xy}, β_x, β_y, χ_x, and χ_y (for the normal incidence of light) of the effective response of the layer of the metamaterial thus introduced can be obtained [46] by parameterizing the scattering matrix

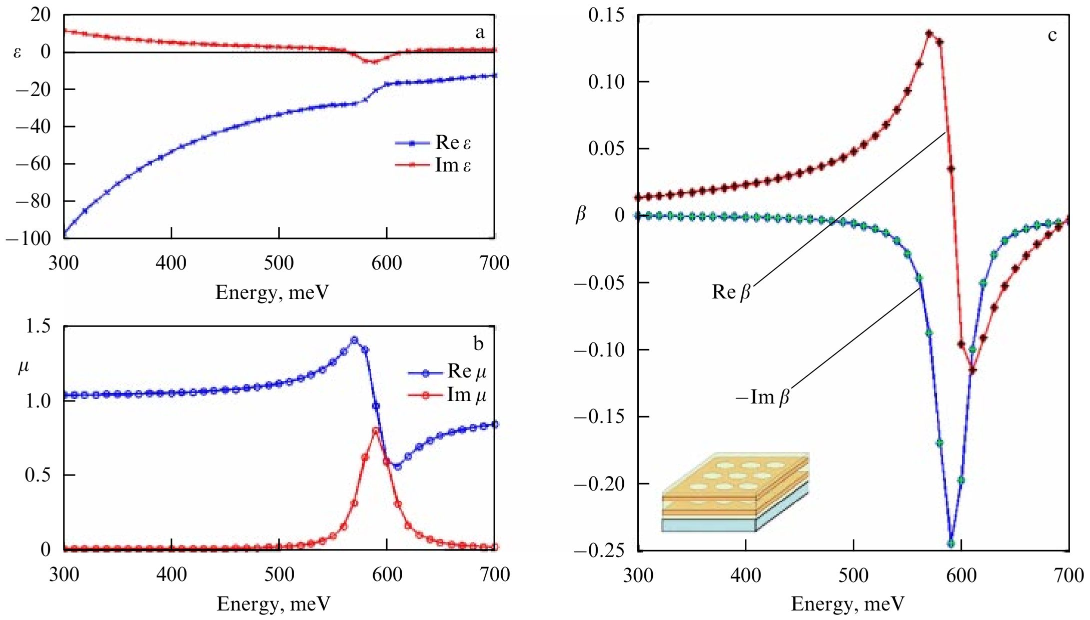

Figure 1. Frequency dependence of the effective electromagnetic response coefficients ε, μ, and β for a metamaterial layer of the bi-fishnet type [28] on a glass substrate ($\varepsilon = 2.32$). The layers represent gold films 10 nm thick perforated by a square lattice of holes (with the period 838 nm and the diameter of holes 360 nm); the films are separated by a dielectric ($\varepsilon = 2.72$) with the thickness 60 nm.

in the far field calculated for this layer [19, 47, 48]. For the normal incidence of light with the wavelength exceeding the period of the metamaterial (such that there is no diffraction, and the layer of the metamaterial behaves like a layer of a uniform material), the scattering matrix in the far field has the size 4×4 and is symmetric ($S_{ij} = S_{ji}$) as a result of the reciprocity of the channels of scattering. Therefore, only 10 of the 16 components of S_{ij} are linearly independent, for example, those with $i \leqslant j$, which exactly corresponds to the number of linearly independent response coefficients in formula (1).

The results of the parameterization of the scattering matrix for metamaterials layers of a bi-fishnet type [28] are illustrated in Fig. 1 for an asymmetric dielectric environment and in Fig. 2 for a symmetric environment. A frequency region is shown near the plasmonic magnetic resonance at $\hbar\omega = 0.6$ eV in which the system has a negative refractive index. Although the layer of the metamaterial by itself has an inversion symmetry, it is seen that in the case of an asymmetric dielectric environment, its effective electromagnetic response has a sufficiently strong resonant bianisotropy (Fig. 1c). But if the layer is located in a symmetric dielectric

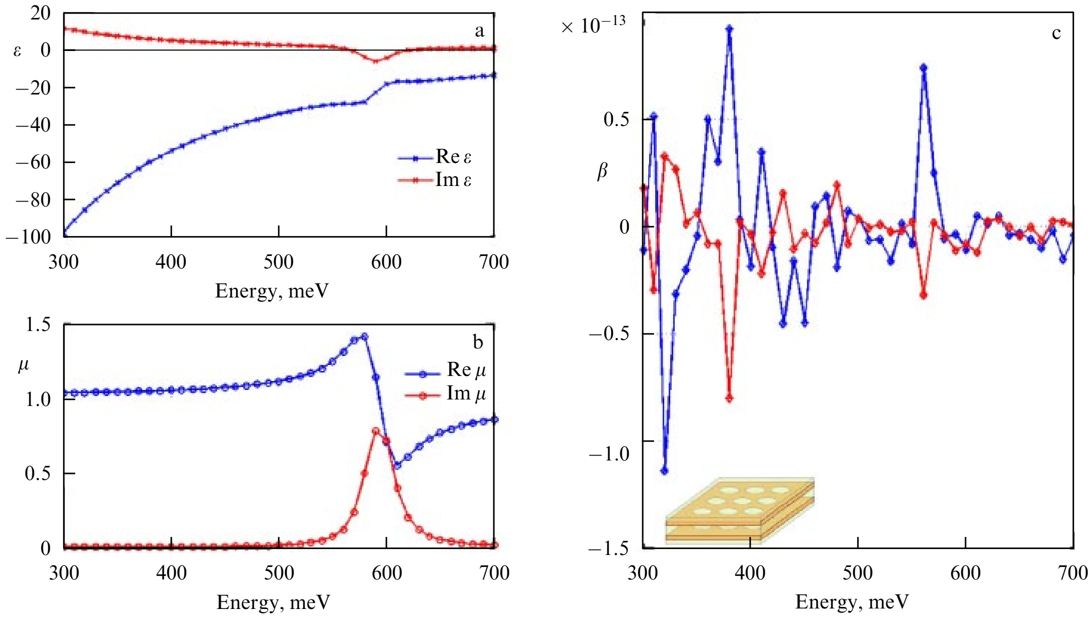

Figure 2. Frequency dependence of the effective electromagnetic response coefficients ε, μ, and β for a metamaterial layer of the bi-fishnet type analogous to the material described in Fig. 1, but in a symmetric dielectric environment (air from above and from below). The bianisotropy coefficient in this case vanishes, which indicates that the structure as a whole is centrally symmetric.

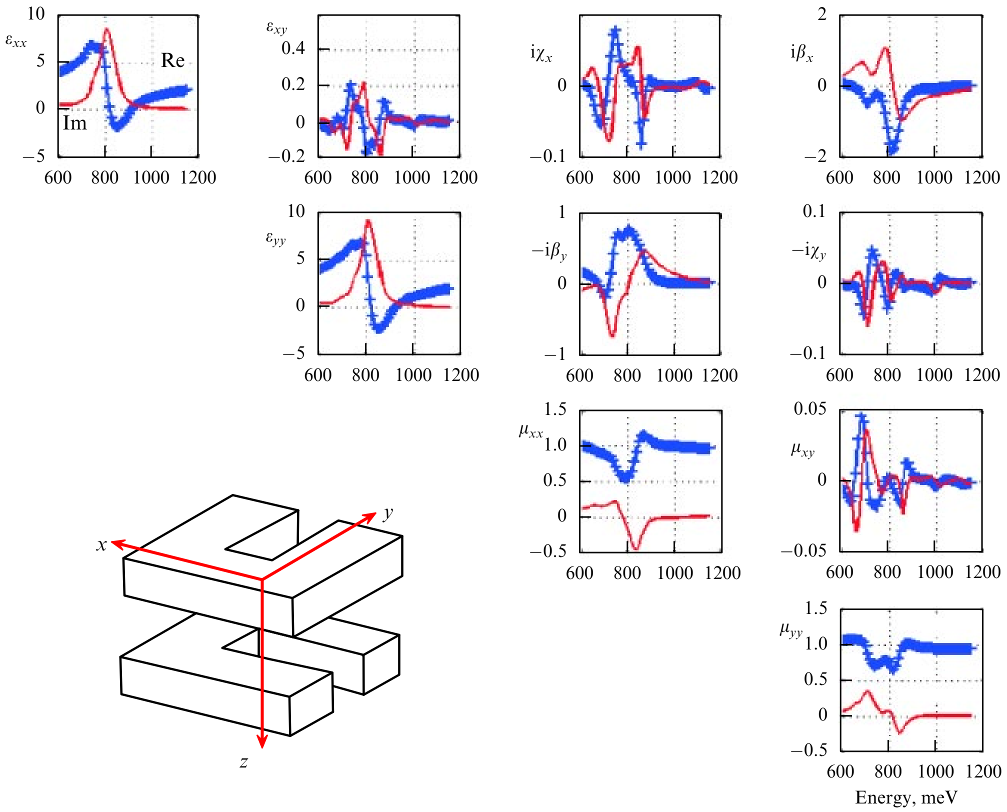

Figure 3. Frequency dependence of the effective coefficients of electromagnetic response $\hat{\eta}_{\|}$ [see Eqn (1)] for a layer of metamaterial in the form of C resonators turned by an angle of 90°. The solid (blue) curves correspond to the real parts of the coefficients, and the thin lines, to the imaginary parts. The unit cell of the bi-layer is shown in the inset. The horizontal dimensions of a C resonator are 230×230 nm^2, the horizontal width of the line is 90 nm, the thickness is 50 nm, the spacing between the layers of the rotated C resonators is 70 nm. The layer of the metamaterial is obtained as a result of horizontal translations of the unit cell with a square lattice of 700×700 nm. The C resonators made of gold are located inside the dielectric layer with $\varepsilon = 2.4$ on the surface of glass ($\varepsilon = 2.25$).

environment (see Fig. 2), the bianisotropy disappears, as in the case of a centrally symmetric structure. These results are the direct demonstration of the nonlocality of the electromagnetic response of thin layers of metamaterials: the effective susceptibilities are not only the characteristics of the structure itself but also depend on the dielectric environment. Therefore, the effective susceptibilities can be used only with great caution, remembering that they are not the characteristics of a bulk metamaterial, but adequately characterize only the response of a specific finite-thickness structure in a given dielectric environment.

Figure 3 shows the results of parameterization of the scattering matrix for a thin layer of a chiral stereometamaterial [31], which has strong natural optical activity. In this case, the system has the complete set of nonzero components of the response matrix $\hat{\eta}_{\|}$.

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Plasmon effects in In(Ga)N-based nanostructures

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In this report, we consider the influence of effects of localized plasmons in metallic nanoparticles on optical processes in In(Ga)N-based structures. The emission and absorption of light and the generation of photoinduced charge carriers is investigated; data on the estimation of the averaged enhancement in InN/In nanocomposites are given.

Plasmonics is a rapidly developing field of applied physics and nanotechnology characterized by the use of effects related to collective oscillations of conduction electrons in metallic structures (plasmons), frequently for quite uncommon applications [1]. The many-sidedness of plasmonics is manifested in various applications such as the realization of media with a negative refractive index [2], the creation of markers used in decoding human genome [3], the enhancement of the luminescence of organic semiconductors [4], an increase in the efficiency of photodetectors [5] and light-emitting diodes [6], controlling liquid-crystal layers [7], and the generation of emission in the terahertz range [8]. Of great interest are also the fundamental properties of plasmonic metastructures, for example, the formation of strong plasmon–polariton resonances, which can be efficiently controlled by changing their structure [9, 10].

There is one additional promising avenue for the realization of the potential of plasmonics: the creation of single-photon sources that can work at room temperature. The local enhancement of an electromagnetic field by plasmons and, correspondingly, an increase in the rate of spontaneous recombination near a metallic surface are analogs of the Purcell effect [11] in microresonators. We note that a reproducible production of microresonators with quantum dots based on wide-gap semiconductors for the same purpose is at present quite problematic.

Here, we consider optical effects in semiconductor layers and nanocomposites caused by localized plasmons (Mie resonances) excited in metallic nanoparticles. The use of particles instead of a continuous film has some advantages [12]. In particular, because of the curvature of the particle surfaces, the interaction of plasmonic excitation and light emission, forbidden due to a difference in their wave vectors, is then allowed. In essence, the study whose results are presented in this report, especially in the part that concerns nanocomposites, is a continuation of the investigation of InN layers with spontaneously formed In clusters [13–19]. In the course of this investigation, we discovered Mie resonances in the spectra of thermally detected optical absorption (TDOA) and established that the plasmons exert a noticeable influence on the emission. For illustration, Fig. 1 depicts combined images obtained via scanning electron microscopy (SEM) and micro-cathodeluminescence (micro-CL) studies of one and the same region of an InN layer. These images show an enhancement of infrared micro-CL near metallic clusters and near pores surrounded by In precipitates.

Current studies are mainly conducted using two systems: (1) InGaN and an Au nanoparticle, and (2) InN with specially formed In clusters. The choice of these pairs of materials is by no means accidental. The energy of plasmonic resonances in Au particles is close to the energy of excitonic transitions in layers of the solid alloy $In_{0.25}Ga_{0.75}$. Indium as a plasmonic metal is less known than gold. But, the applicability of a metal for the amplification of optical processes is determined by the oscillator strength of plasma resonance. The spectral dependence of this parameter can be described by the ratio of the real part of the complex dielectric function of metal to its imaginary part, $|\mathrm{Re}\,\varepsilon|/\mathrm{Im}\,\varepsilon$. In the infrared range (0.7–1 eV), where radiative optical transitions are observed in InN, this ratio for In is not less than that for Au in the visible range (about 2 eV) (Fig. 2).

An increase in the efficiency of emission implies the formation of coupled localized plasmon–radiating dipole states. For the radiative plasmon decay, the metallic particle can act as a radiating antenna; but in the case of the dominance of a nonradiative decay (with large internal losses of the particle), the coupling can quench emission of the radiating dipole. The balance between these components depends, among other factors, on the particle size [20]. In the general case, to guarantee the interaction of the radiating dipole and localized plasmon, a number of conditions must be

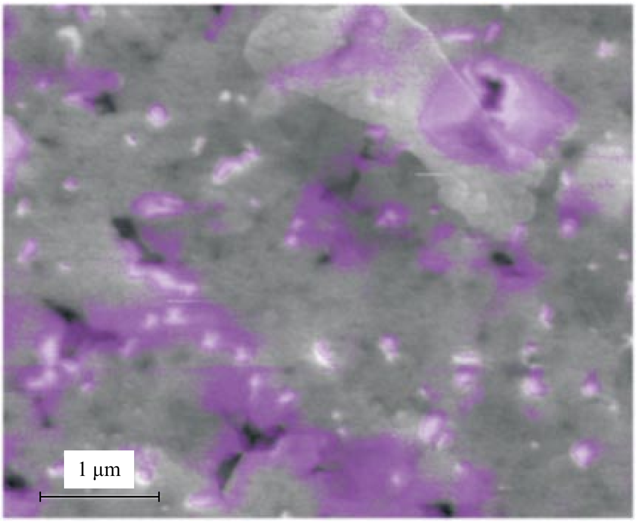

Figure 1. Superimposed images of the same region of a layer of InN obtained by SEM and micro-CL. The clusters and pores are respectively seen as bright and dark spots. The violet spots (in the electronic version of the paper at http://www.ufn.ru) correspond to intense infrared (0.75 eV) cathodoluminescence.