

## Research article

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# Two optical sensing elements for H<sub>2</sub>O and NO<sub>2</sub> gas sensing based on the single plasmonic – photonic crystal slab

<https://doi.org/10.1515/aot-2019-0059>

Received November 13, 2019; accepted May 27, 2020;

published online July 20, 2020

**Abstract:** Two optical sensing elements based on the surface plasmon waves at the plasmonic-photonic-crystal/air interface, excited in the Kretschmann configuration, are proposed. The sensing elements are designed to detect air humidity and NO<sub>2</sub> concentration in air. The angular reflectance spectra of the sensing elements are theoretically analyzed as the function of the analyte concentration. The proposed NO<sub>2</sub>-sensing element has no cross-sensitivity to humidity. The two sensing elements are based on the same multilayer metal-dielectric structure with the only exception on different gas-sensitive material layers. When combined, the sensing elements can be used to measure humidity and NO<sub>2</sub> concentration in humid air.

**Keywords:** optical humidity sensor; optical nitrogen dioxide sensor; photonic crystal surface wave; sensor selectivity.

## 1 Introduction

The phenomena of the surface wave excitation on a photonic-crystal/dielectric interface [1, 2] and surface plasmonic resonance [3, 4] have long been used for biological and chemical sensing. Both approaches have

advantages and disadvantages. For example, sensing elements based on plasmonic resonance have only one layer of a noble metal (Ag, Au, or sometimes Cu, see [5]) coated with a functional (analyte-sensitive) layer. Exposure to an external chemical or biological agent leads to a change in the refractive index of the functional layer, and, therefore, to a spectral shift of the plasmonic resonance, easily detected from the reflection coefficient in the Kretschmann configuration [3, 4]. Such sensing elements are quite easy to fabricate (they consist of only a few material layers). However, owing to ohmic losses in the metal, surface plasmon resonance has relatively low Q-factor, which limits sensitivity of the sensors based on it.

Sensing elements based on photonic crystals (PhCs) operate in a similar way: a functional layer sensitive to exposition to an external chemical or biological agent is deposited onto the surface of a (usually one-dimensional) photonic crystal [1]. The functional layer's refractive index change leads to a spectral shift of the surface wave propagating along the PhC/functional layer/air interface, which is registered optically. One could think that in such photonic crystals-based sensing elements, an arbitrarily large Q-factor of the surface-wave excitation and therefore large sensitivity is reachable. However, to observe the resonance associated with the surface wave excitation (higher Q than the plasmonic resonance) in the common Kretschmann configuration, it is necessary to introduce carefully matched absorption (or, equivalently, scattering by inhomogeneities) into the structure. Thus, in the absence of absorption/scattering, the reflection coefficient is 1, and on the other hand, the resonance will be low Q if the absorption is too large. Recently [6], a sensing element based on surface waves in magneto-optical materials was proposed to overcome this limitation. In the study by Merzlikin et al [6], it was shown that the enhancement of Kerr polarization rotation (caused by the excitation of a surface wave) can be used to indicate the surface wave spectral shift due to exposure to analytes. Moreover diminishing absorption results in increasing of Kerr polarization rotation. For surface-plasmon magneto-optic sensors, see also the studies by Rizal et al [7] and Rizal and Belotelov [8].

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Currently, there is a tendency to combine the advantages of both approaches (using both plasmonic and photonic-crystal surface waves) in structures with a PhC coated with a thin layer of noble metal [9–11]. In this article, we consider two optical sensing elements based on surface waves in a plasmon-PhC structure excited in the Kretschmann configuration. The proposed sensing elements are designed to detect air humidity and nitrogen dioxide (NO<sub>2</sub>) concentration in air (including humid air).

## 2 Humidity-sensing element

The proposed humidity-sensing element is based on a refractive index change of the humidity-sensitive polymer Nafion. In this sensing element, the angular reflectance spectrum in the Kretschmann configuration changes depending on the Nafion refractive index [12].

The sensing element is a layered system [Ta<sub>2</sub>O<sub>5</sub>/SiO<sub>2</sub>]<sub>10</sub>/Au/SiO<sub>2</sub>/Nafion/air, i.e., on a PhC with 10 two-layer periods formed by Ta<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub> layers, an Au, a SiO<sub>2</sub>, and, finally, Nafion layers are subsequently placed (see Figure 1(a)). The analyzed humid air is above the entire multilayer system. The thicknesses of Ta<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub>, Au, and Nafion layers are 135, 207, 10, and 1000 nm, respectively. The thickness of the SiO<sub>2</sub> layer above Au is 20 nm. All of these thicknesses are optimized for the working wavelength of 658 nm. The SiO<sub>2</sub> layer upon the Au layer plays two roles. First, it isolates the vulnerable thin metal layer from the environment and extends the sensing element life term (this is especially important if less chemically inert Ag is used instead of Au). Second, this SiO<sub>2</sub> layer increases the element sensitivity to the Nafion refractive index change [9]. The layers thicknesses listed earlier were obtained from numerical optimization: the thicknesses of the Ta<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub> layers of the PhC and the thickness of the SiO<sub>2</sub> layer upon Au were varied to maximize the sensitivity – the power-reflectance change per unit Nafion refractive index change. Only the *p*-polarized incident wave was considered. During the optimization, we considered only those PhC layers' thicknesses for which the PhC had a bandgap at incidence angles larger than the total reflection angle in air (i.e., when the exciting wave wavevector component parallel to the layers is less than  $k_0$ ). The bandgap existence is necessary to a surface wave existence at the PhC/Au/SiO<sub>2</sub>/Nafion/air interface. For the optimization, we took the Ta<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub>, and Au refractive indices as  $n_{\text{Ta}_2\text{O}_5} = 2.131$ ,  $n_{\text{SiO}_2} = 1.456$ , and  $n_{\text{Au}} = 0.143 + 3.682i$ , respectively (at  $\lambda = 658$  nm, [13–15]). The Nafion refractive index was taken to be  $n_{\text{Naf}} = 1.358$  (which corresponds to the relative humidity [RH] of 50% [16]). During

optimization, a thinner Au layer usually resulted in larger maximum sensitivity corresponding to the surface wave excitation. That is why we chose the thinnest possible Au layer of 10 nm which is technologically reproducible and solid.

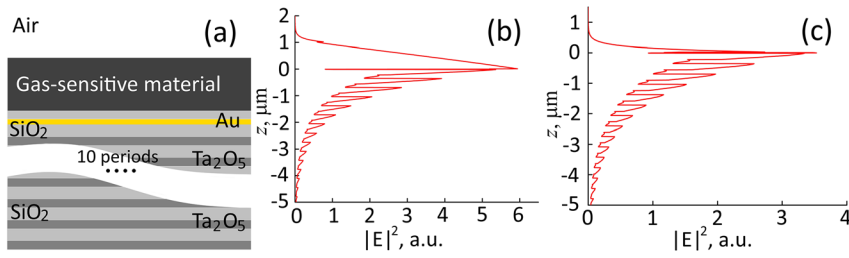
The exciting wave incidence angles given in the following correspond to the incidence (from the PhC side to the Nafion/air interface) angles in a prism made of BK7 glass with the refractive index of 1.51 (at  $\lambda = 658$  nm, [17]).

Figure 2(a,b) shows the calculated angular power-reflectance spectrum for a *p*-polarized incident wave in the Kretschmann configuration (plane wave incidence from a BK7 prism onto the multilayer sensing element system). The narrow dip near the angle of 1.116 rad corresponds to the excitation of a surface wave. In Figure 1(b), the field distribution of the surface wave excited is shown. When the RH changes from 50 to 20% (temperature is about 24° C), the dip shifts by 0.010 rad toward the smaller angles (the refractive index of Nafion decreases from 1.358 to 1.346 [16]). The corresponding maximum change in power reflectance is 97%. The calculated maximum sensitivity is 1730 1/RIU (change in power reflectance per unit Nafion refractive index change) at the RH of 50% (see Figure 2(c)) and 1470 1/RIU at the RH of 20%. In the linear approximation of the Nafion refractive index variation with the RH variation, we get the sensing element sensitivity of 0.69 1/RH% (i.e., for the RH change by 1% the reflectivity changes by 69%) around the RH of 50%.

In addition to the sensitivity to humidity, we considered the cross-sensitivity to NO<sub>2</sub> in air for the humidity-sensing element. Because there are no data on the effect of NO<sub>2</sub> on the Nafion optical properties, only a change in the air refractive index under the varying NO<sub>2</sub> concentration was considered. When the NO<sub>2</sub> concentration changes from 0 to 100 ppm, the dielectric constant of air in the visible range increases by  $10^{-7}$  [18]. In Figure 2(d), the absolute value of the power-reflectance change upon the NO<sub>2</sub> concentration change by 100 ppm is shown. Based on these calculations, we see that the humidity sensor sensitivity to NO<sub>2</sub> due to air refractive index change is significantly lower than the sensitivity of the proposed NO<sub>2</sub>-sensing element described in the following.

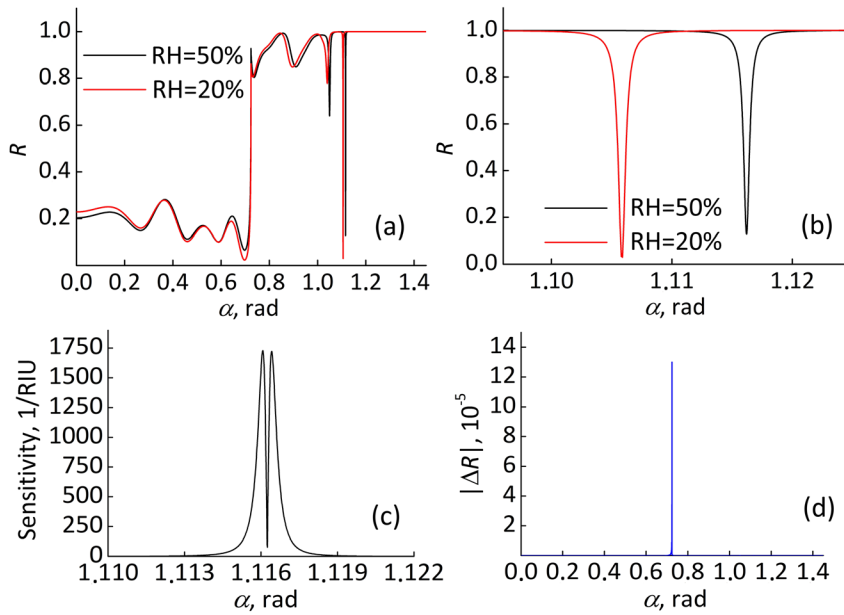
## 3 NO<sub>2</sub>-sensing element

The proposed NO<sub>2</sub>-sensing element is based on a refractive index change of the gas-sensitive material – Pb-phthalocyanine – depending on the concentration of NO<sub>2</sub> in air near the sensing element. In this case, the angular



**Figure 1:** (a) The schematic of the sensing elements of humidity and NO<sub>2</sub> composed of a photonic crystal slab with Au, SiO<sub>2</sub>, and gas-sensitive layers atop. Light-gray layers are SiO<sub>2</sub>, gray layers are Ta<sub>2</sub>O<sub>5</sub>, the yellow layer is Au, and the dark-gray layer is gas-sensitive material (either Nafion or Pb-phthalocyanine). The layer thicknesses are not in scale. Below this multilayer structure, there is a BK7 prism. (b) and (c) The electric

field norm of the surface wave which is excited for sensing in the case of the humidity- and NO<sub>2</sub>-sensing element, respectively. The  $z$  coordinate is along the normal to layers, and  $z = 0$  corresponds to the top Au layer interface.



**Figure 2:** (a) and (b) Angular power-reflectance spectra of the  $p$ -polarized plane wave for the humidity-sensing element. The black curve is for the RH of 50%, and the red curve is for the RH of 20%. Part (b) is the enlarged view of part (a). (c) The sensitivity of the humidity-sensing element to Nafion refractive index variation at the RH of 50% as a function of the exciting wave incidence angle. (d) The absolute value of the power-reflectance change  $\Delta R$  upon the NO<sub>2</sub> concentration change by 100 ppm due to air refractive index change at a constant humidity is shown for the humidity-sensing element. The wavelength is  $\lambda = 658$  nm. RH, relative humidity.

reflectance spectrum of the sensing element in the Kretschmann configuration changes depending on the Pb-phthalocyanine refractive index.

The sensing element is a layered system [Ta<sub>2</sub>O<sub>5</sub>/SiO<sub>2</sub>]<sub>10</sub>/Au/SiO<sub>2</sub>/Pb-phthalocyanine/(air+NO<sub>2</sub>), i.e., on a PhC with 10 two-layer periods formed by the layers of Ta<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub>, an Au layer is placed, then a SiO<sub>2</sub> layer and, finally, a Pb-phthalocyanine layer (see Figure 1(a)). The analyzed air with NO<sub>2</sub> is above the entire multilayer system. We considered the same Ta<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub>, and Au layers' thicknesses as were determined for the humidity-sensing element in the previous section. The optimization of the Pb-phthalocyanine layer thickness gave the value of 20 nm. This optimization was performed for the Pb-phthalocyanine refractive index of  $n_{\text{PhthC}} = 3.684 + 1.712i$  (which corresponds to zero NO<sub>2</sub> concentration [19]). The full optimization with respect to Ta<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub>, and Au layers' thicknesses did not give the significantly larger sensitivity to NO<sub>2</sub> compared with that for the layers' thicknesses listed in the previous section. As in the case of

the humidity-sensing element, the incidence angles of the exciting wave given in the following correspond to the incidence angles in a prism made of BK7 glass.

In Figure 3(a,b), the angular power-reflectance spectrum for a  $p$ -polarized incident wave in the Kretschmann configuration is shown for the NO<sub>2</sub>-sensing element. The dip near the angle of 1.02 rad corresponds to the excitation of a surface wave. In Figure 1(c), the field distribution of the surface wave excited is shown. When the NO<sub>2</sub> concentration changes from 0 to 100 ppm, this dip shifts by 0.0008 rad toward the larger angles and becomes more shallow (the refractive index of Pb-phthalocyanine changes from  $3.684 + 1.712i$  to  $4.88 + 1.22i$  [19]). The corresponding maximum change in reflection coefficient is 0.32. The calculated maximum sensitivity is 0.24 1/RIU (change in power reflectance per unit change of the Pb-phthalocyanine refractive index real part) at the zero NO<sub>2</sub> concentration (see Figure 3(c)) and 0.23 1/RIU at the 100 ppm NO<sub>2</sub> concentration. In the linear approximation of the Pb-phthalocyanine

refractive index variation with the NO<sub>2</sub> concentration variation, we get the sensing element sensitivity of 0.0029 1/ppm around the zero NO<sub>2</sub> concentration.

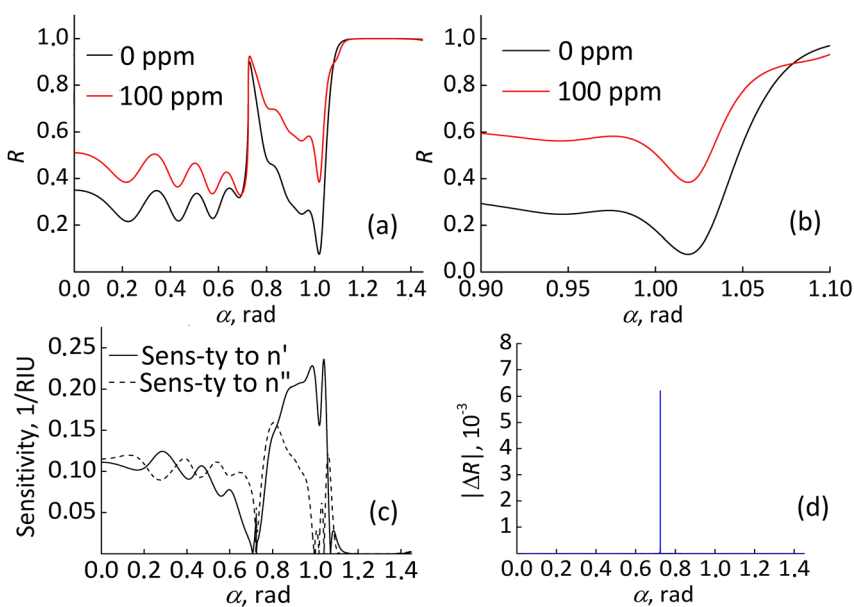
The cross-sensitivity of the NO<sub>2</sub>-sensing element to humidity was analyzed. According to Hamann et al [20], Pb-phthalocyanine is not optically sensitive to air humidity. Therefore, only a change in the refractive index of air upon the changing humidity was considered. In accordance with the experimental data from the study by Mathar [21], an increase in RH from 0 to 100% leads to a decrease in the air dielectric constant by  $2 \cdot 10^{-6}$ . In Figure 3(d), the absolute value of the power-reflectance change upon the RH change from 0 to 100% is shown. From these calculations, the cross-sensitivity of the NO<sub>2</sub>-sensing element to humidity is significantly lower than the sensitivity of the proposed humidity sensor described earlier.

## 4 Discussion and conclusions

Using the excitation of the surface wave on a PhC surface can increase sensor sensitivity compared with that of a sensor based on a surface plasmon excitation on a thick metal film surface. To confirm, we optimized the sensitivity of a sensing element based on the multilayer system Au/Nafion/air as a humidity-sensing element. In such a system, the surface plasmon is excited at the Au/Nafion/air interface in the Kretschmann configuration for the *p*-polarized exciting wave. The optimization was performed with respect to the Au and Nafion layers thicknesses at  $\lambda = 658$  nm. The maximum sensitivity to humidity for the Nafion layer thickness

$\leq 1000$  nm was 660 1/RIU and was achieved at the Au layer thickness of 60 nm and the Nafion layer thickness of 1000 nm. When thinner Nafion layers  $\leq 300$  nm are considered, the maximum sensitivity was 58 1/RIU (achieved at the Au layer thickness of 55 nm and the Nafion layer thickness of 300 nm) for the surface plasmon-based sensing element. The optimization of a [Ta<sub>2</sub>O<sub>5</sub>/SiO<sub>2</sub>]<sub>10</sub>/Au/SiO<sub>2</sub>/Nafion/air structure for Nafion thickness  $\leq 300$  nm gives the sensitivity to humidity up to 602 1/RIU (achieved at the Ta<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub>, Au, and Nafion layers thicknesses of 114, 253, 10, and 300 nm, respectively, and the thickness of the SiO<sub>2</sub> layer above Au of 30 nm). We believe that the considerably larger sensitivity of the PhC-based sensitive element than that of surface plasmon-based sensitive elements is due to the larger ohmic losses of a surface plasmon in the thicker Au films. On the other hand, some losses (either ohmic or due to scattering on roughness) are needed for the reflectance to have resonant features and not to be trivial 1.0 at incidence angles larger than the total internal reflection angle. The thin Au layer plays the role of such a lossy layer in our humidity-sensing element.

Nafion polymer has an inhomogeneous porous structure which leads to optical scattering in it. Therefore, it is interesting to evaluate the optical losses due to scattering in Nafion, to compare them with the ohmic losses in the Au layer and to decide whether it is sufficient to remove the Au layer and to rely only on the scattering losses in Nafion for sensing. For this purpose, we evaluated the imaginary part of Nafion effective dielectric permittivity describing scattering losses from the Nafion structure characteristics. According to Lehmani et al [22], the characteristic porosity (the volume of pores over the total volume of porous



**Figure 3:** (a) and (b) Angular power-reflectance spectra of the *p*-polarized plane wave for the NO<sub>2</sub>-sensing element. The black curve is for the zero NO<sub>2</sub> concentration, and the red curve is for the 100 ppm NO<sub>2</sub> concentration. Part (b) is the enlarged view of part (a). (c) The solid line and the dashed line indicate the sensitivities of the NO<sub>2</sub>-sensing element to Pb-phthalocyanine refractive index real part and imaginary part variation, respectively, at the zero-NO<sub>2</sub> concentration as the function of the exciting wave incidence angle. (d) The power-reflectance change  $\Delta R$  upon the RH change from 0 to 100% at the zero NO<sub>2</sub> concentration is shown for the NO<sub>2</sub>-sensing element. The wavelength is  $\lambda = 658$  nm. RH, relative humidity.

polymer) of Nafion 117 (used in humidity sensors [12]) is about 0.2–0.3 and the pore diameter is about 1–5 nm. For the case of porosity of 0.3 and pores with diameter of 5 nm filled with air (more optically contrast than pores filled with water), we evaluated the effective dielectric permittivity imaginary part in the Rayleigh scattering limit according to Tsang and Kong [23]. The determined scattering-related Nafion effective dielectric permittivity imaginary part is  $\varepsilon''_{\text{Naf}} = 7 \cdot 10^{-6}$ . Such a value of loss added to Nafion leads to the maximum change of 24 1/RIU for our humidity-sensing element sensitivity. Compared with the sensitivity of 1730 1/RIU, this change can be considered negligible. Thus, we conclude that the scattering in Nafion is rather small and the thin Au layer is necessary.

In the case of the NO<sub>2</sub>-sensing element, the Pb-phthalocyanine layer is rather lossy and the Au layer is not necessary. In addition, we considered the layered system Au/Pb-phthalocyanine/air as a surface plasmon-based NO<sub>2</sub>-sensing element and optimized it with respect to the Au and the Pb-phthalocyanine layers thicknesses. The maximum achieved sensitivity to NO<sub>2</sub> is 0.23 1/RIU (realized at the Au and Pb-phthalocyanine layers thicknesses of 15 and 20 nm, respectively), which is nearly the same as for our PhC-based sensing element. Thus, we conclude that for such a lossy material as Pb-phthalocyanine, the sensitivity is primarily determined by the Pb-phthalocyanine layer properties and weakly depends on losses in other components of a layered system. Nevertheless, in our proposed NO<sub>2</sub>-sensing element, we use the PhC-based layered system because it is the same as in the proposed humidity-sensing element and the two sensing elements fabricated on the common platform could be used in combination with each other.

In summary, two optical sensing elements based on a photonic-plasmonic crystal with the same layered structure in the Kretschmann configuration were proposed. The sensing elements differ from each other by only one top gas-sensitive material layer. One of the sensing elements is designed to detect air humidity and the second one, to detect NO<sub>2</sub> concentration in air. Typically, when detecting the concentration of various gases in a mixture, a cross-sensitivity problem arises. In many cases, this problem arises owing to cross-sensitivity to air humidity when measuring concentration of some other gases [24, 25]. We proposed the NO<sub>2</sub>-sensing element in which Pb-phthalocyanine is used as gas-sensitive material. According to Hamann et al [20], Pb-phthalocyanine has no cross-sensitivity to humidity. The sensitivity of the proposed NO<sub>2</sub>-sensing element to air refractive index change due to RH variation was shown to be negligible. Thus, we assume that the NO<sub>2</sub>-sensing element could be used to detect NO<sub>2</sub>

concentration without cross-sensitivity to humidity. In the proposed humidity-sensing element, Nafion polymer is used as the humidity-sensitive material. This polymer is porous, and its sensitivity to humidity is based on filling pores with water and swelling [26]. Unfortunately, there are no data on the NO<sub>2</sub> effect on the Nafion optical properties. We showed that the humidity-sensing element sensitivity to air refractive index variation due to NO<sub>2</sub> concentration variation is negligible compared with that of the NO<sub>2</sub>-sensing element. Nevertheless, RH could also be measured using the humidity-sensing element, taking into account the known NO<sub>2</sub> concentration (determined by the NO<sub>2</sub>-sensing element) and a cross-sensitivity of the humidity-sensing element to NO<sub>2</sub>, if any. Thus, the proposed sensing elements in combination with each other (e.g., on the single multilayer substrate) could be used to measure humidity and NO<sub>2</sub> concentration in humid air mixed with NO<sub>2</sub>.

**Acknowledgments:** This work was supported by Russian Foundation for Basic Research, project 18-29-24204.

**Author contribution:** A.M. Merzlikin is the author of the idea and supervisor of the work. He participated in preparation of the manuscript. A.I. Ignatov made optimization of the structures and calculations of the spectra. He also participated in preparation of the manuscript.

**Research funding:** This work was supported by Russian Foundation for Basic Research, project 18-29-24204.

**Conflict of interest statement:** The authors declare no conflicts of interest regarding this article.

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