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Laser adaptive holographic system for microweighing of nanoobjects

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Abstract. **A system for measuring the mass of micro- and nanoobjects based on resonance microweighing using the principles of adaptive holographic interferometry is proposed and experimentally implemented. The sensitive element of the system is a microcantilever to which the objects to be weighted are attached. The eigenoscillations of the microcantilever are excited with a laser pulse. The detection of oscillations is implemented using the adaptive holographic interferometer, the key element of which, the dynamic hologram, is formed in the photorefractive crystal CdTe. The detected variation in mass of the particles, attached to the microcantilever, amounted to $(420 \pm 9) \times 10^{-12}$ g, the measurement error being 8.5×10^{-12} g. The sensitivity of the measurement system is 1.7×10^{-12} Hz g^{-1}. The possibility of increasing the sensitivity of the system by 6.5×10^6 times and reducing the mass detection threshold by 1.5×10^7 times by microcantilevers of submicron size is experimentally demonstrated.**

Keywords: *resonance microweighing, microcantilever, adaptive interferometry.*

1. Introduction

Measuring ultra-small masses is an important component of studies in the field of nanotechnologies, biology, chemistry, material science, rheology, etc. [1–3]. Traditionally the masses of atoms and molecules are measured using the mass spectrometry method [4]. However, the application of this method to measuring the masses of complex protein compounds or nanoparticles appears to be extremely difficult and in some cases practically impossible, since the measurement of the electric charge in such objects is a separate complex problem [5]. From this point of view, a more preferable approach to mass determination in micro-objects is the resonant microweighing that consists in measuring the eigenoscillation frequency of oscillatory systems – micro-oscillators, to which the micro- or nanoobjects are attached [2, 6]. This approach is used to determine the masses of small particles, including protein molecules, viruses and bacteria [1, 6–9]. The detection of micro-object and micro-oscillator oscillations is implemented mainly using optical methods [1–3, 8, 9], among which the most sensitive are interferometric methods that potentially allow detection of oscillations with the amplitude of a few angstroms.

However, the use of a classical interferometer in such systems is associated with a number of technical difficulties. The first of them is caused by the high sensitivity of the interferometer that makes it strongly influenced by the external factors (variation of temperature, pressure, uncontrollable deformations, seismic microvibrations, etc.), which violates the measuring system operation stability, gives rise to additional noises and, therefore, increases the detectable mass threshold. The use of electronic systems to stabilise the interferometer operating point essentially complicates the whole measuring system and introduces additional noises into its operation [10].

Another key problem that hampers the design of resonance microweighing systems on the basis of interferometers is caused by the fact that the increase in their sensitivity is inseparably linked with the necessity to reduce the microoscillator size. The sizes of modern microoscillators are comparable with the transverse dimensions of the probe (object) laser beam in the focus. Their further miniaturisation leads to significant distortions of the object wave front in the interferometer. In turn, the mismatch of the wave fronts of the interfering beams leads to the reduction of the interferometer sensitivity up to the total failure of its operation. The mentioned problem becomes particularly urgent in the case of using the oscillators of submicron and nanometre scale. Moreover, due to technological specificity the microoscillator surface cannot be always manufactured mirror-smooth, which also introduces additional wave front distortions, reduces the precision of mass measurement and increases the detection threshold.

A complex solution to the above problems consists, apparently, in constructing ultra-small mass measuring systems on the basis of adaptive interferometry principles [11, 12]. The key component of an adaptive interferometer is a dynamic hologram, produced in a photorefractive crystal (PRC). The holographic principle of overlapping the waves in the crystal provides precise matching of the wave fronts in the reference and the object light beams, and the adaptive properties of the dynamic hologram provide stabilisation of the interferometer operating point in the region of maximal sensitivity, where the quadrature conditions are realised. The latter eliminates the influence of uncontrollable variations of the environmental parameters [13], making it possible to use such systems under the out-of-lab conditions. Thus, the design of

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systems based on dynamic holograms recorded in PRCs promises an essential increase in their sensitivity at the expense of using extremely small microoscillators of the arbitrary shape and with the arbitrary surface structure.

In this connection, the aim of the present work was to construct and practically approve a system for measuring ultra-small masses on the basis of resonance microweighing with the use of the adaptive holographic interferometry principles.

2. Experimental setup

In the present study the role of a sensitive element (microoscillator) was played by the microcantilever with the dimensions 180 × 40 × 15 μm made of silicon. One of the ends of the microcantilever was fixed on a rigid base, the other end left free. The objects to weigh were attached to the free end. The frequency of eigenoscillations of the microcantilever is determined by its geometry, physical parameters, the mass of the attached object, and can be calculated using the following expression [7]

$$f = \frac{\eta^2 T}{2\pi L^2}\left[\frac{E}{12\rho(1+4\gamma)}\right]^{1/2}, \qquad (1)$$

where γ is the ratio of the mass m, attached to the microcantilever, to the microcantilever own mass m_0; ρ is the microcantilever material density; L and T are the length and the thickness of the microcantilever, respectively; E is the Young modulus of the microcantilever material; and η is the dimensionless parameter, depending on γ, $\eta(\gamma) = 1.875/(1 + 4\gamma)$. Note, that in a rather wide interval of γ values (from 0 to 0.01) the parameter η changes only a little (from 1.875 to 1.803) and, therefore, in the calculations it was assumed constant.

The scheme of the adaptive system for measuring small masses is presented in Fig. 1. Free oscillations of the microcantilever are excited by pulses of laser radiation with the wavelength $\lambda = 532$ nm, duration 7 ns, energy 0.5mJ, and repetition rate 1 Hz. The detection of free cantilever oscillations is implemented using the adaptive interferometer. In the latter case, the cw laser producing the radiation with $\lambda = 1064$ nm and the power 20 mW is used as a source. The radiation from the laser is divided into the object and the reference beam with the intensity ratio 1 : 5. The object beam is focused onto the free end of the microcantilever by means a short-focus lens, and after reflection from it arrives through the face (100) of the photorefractive crystal CdTe, where, interacting with the reference beam that propagates along the [010] crystal axis, it produces the dynamic hologram. The interaction of the reference and the object beam of the interferometer at the dynamic hologram provides a transformation of the phase changes, caused by the oscillations of the microcantilever, into the intensity changes [13] registered by a photodetector. Using the technique described in Ref. [14], the sensitivity of the interferometer was found to be 5.2×10^{-8} rad W$^{-1/2}$ Hz$^{-1/2}$. For the intensity 13 W cm^{-2} the time of recording the hologram was 2.5 ms, which corresponds to the cutoff frequency 400 Hz. Hence, the use of the dynamic hologram in the measuring system allows elimination of the noise effect with the characteristic frequency up to 400 Hz. Therefore, the use of the dynamic hologram in the measuring system allows elimination of nose influence with the characteristic frequency up to 400 Hz.

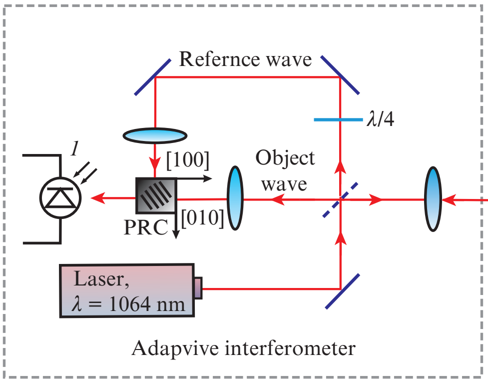


Figure 1. Schematic diagram of the experimental setup for measuring ultra-small masses: (*1*) photodetector; (*2*) microcantilever; (*3*) laser pulse ($\lambda = 532$ nm); (PRC) photorefractive crystal; ($\lambda/4$) quarter-wave plate.

3. Experimental technique and results

The registration of the signal, received by the photodetector, is implemented using the oscilloscope, synchronised with the pulsed laser. From the oscilloscope the data are transferred to the computer used to determine the frequency of the microcantilever oscillations. A typical view of the recorded signal and its Fourier spectrum are presented in Fig. 2. In the oscillogram at the moment of time $t = 0$ s a laser pulse arrives at the microcantilever and excites its oscillation. Then the free

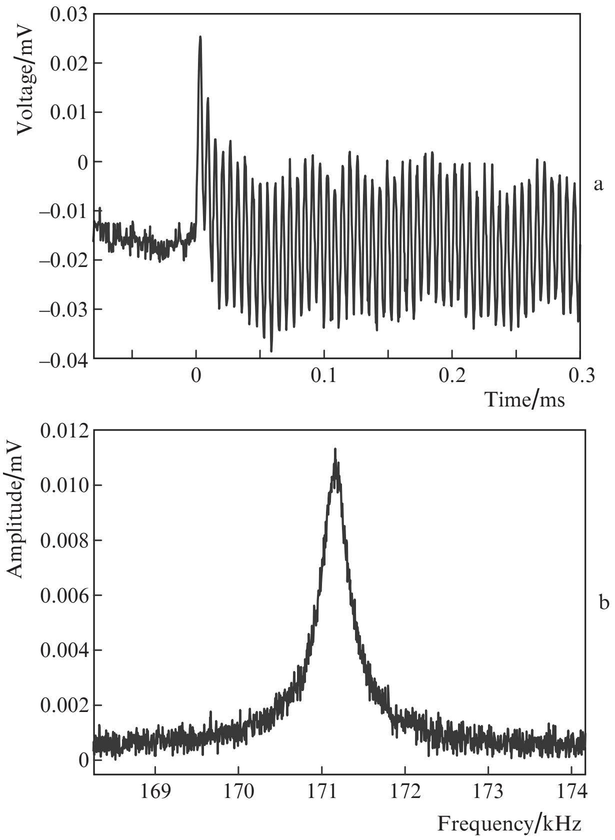


Figure 2. Oscillogram of the signal received by the photodetector (a) and its Fourier spectrum (b).

oscillation begins and lasts during 0.5 ms for the cantilever used in the experiment and the initial energy of the laser pulse 0.5 mJ.

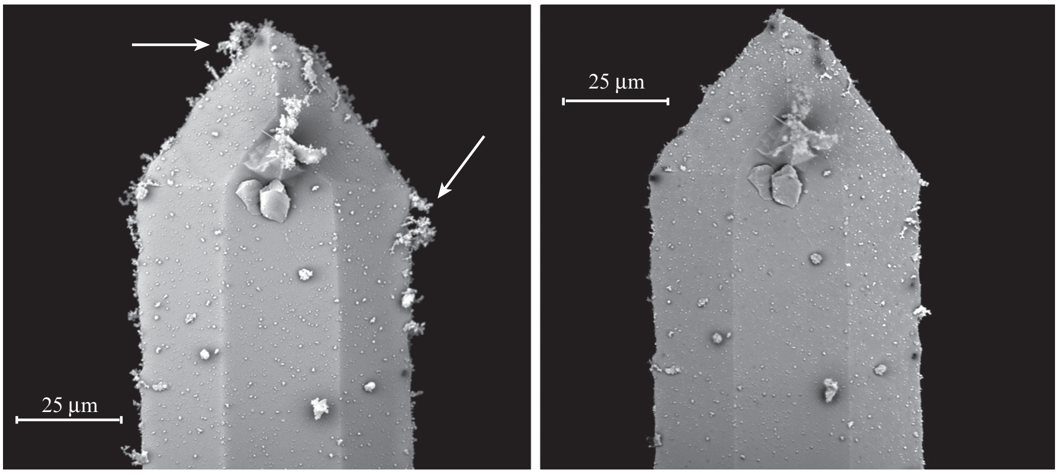
The measuring system was experimentally approved by measuring the mass of nanoobjects, represented by platinum nanoparticles. Before the measurement the nanoparticles with characteristic transverse dimensions from 60 to 400 nm were attached to the microcantilever using the method of magnetron vacuum deposition. In the process of measurement each laser pulse exciting oscillations caused detachment of a certain number of nanoparticles from the microcantilever, which is illustrated by SEM images before and after the action of 62 laser pulses (Fig. 3). The decrease in the mass attached to the cantilever due to gradual removal of nanoparticles caused appropriate growth of the eigenoscillation frequency (Fig. 4). It is seen, that after the action of 62 laser pulses the microcantilever oscillation frequency has grown by 751 Hz. Using the obtained frequency change and Eqn (1), one can determine the total change in the mass of attached particles, which was found to amount to 420×10^{-12} g.

4. Sensitivity of the measuring system and mass detection threshold

From Eqn (1) it is possible to derive the relation between the error δf of frequency measurement and the error δm of mass determination:

$$\delta m = \frac{m_0}{2} \frac{\delta f}{f} . \tag{2}$$

In fact, Eqn (2) determines the threshold of detecting the mass, attached to the microcantilever, which, in turn, is determined by the relative error of frequency measurement. Figure 5 illustrates the statistical dispersion of the eigenoscillation frequencies, experimentally obtained in 16 measurements, performed with the constant mass of the microcantilever m_0 (in the absence of attached mass, $m = 0$), from which the frequency measurement error was determined as $\delta f = 15$ Hz. Hence, for the microcantilever used in the present work the mass measurement error appeared to be 8.5×10^{-12} g.



a b

Figure 3. SEM images of the free end of the microcantilever with nanoparticles attached to it (pointed by arrows) before the laser pulse action (a) and after 62 laser pulses (b).

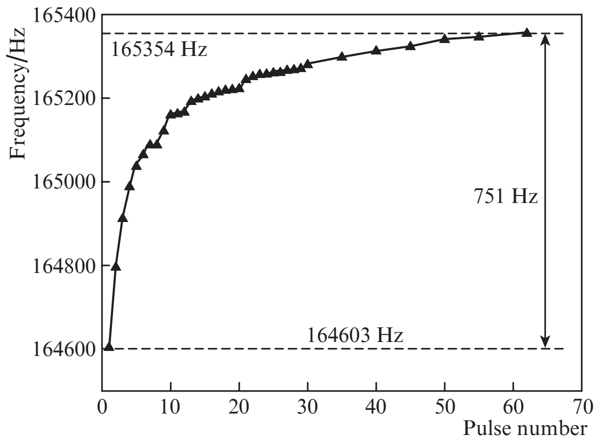


Figure 4. Dynamics of variation of the eigenoscillation frequency for the microcantilever with the attached mass during the action of 62 laser pulses.

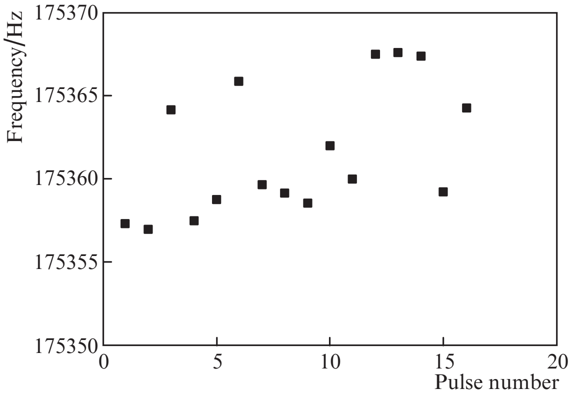


Figure 5. Microcantilever eigenoscillation frequency dispersion.

The sensitivity S of the presented measuring system is defined as the ratio of the oscillation frequency shift and the change of the attached mass: $S = \partial f / \partial m$. Provided that the ratio of the attached mass to the mass of the microcantilever is small ($m/m_0 \ll 1$), the sensitivity S can be expressed in terms of the microcantilever parameters in the following way:

$$S = -a/(WL^3), \tag{3}$$

where $a = (3/\pi^3)[E/(12\rho)^3]^{1/2}$ and W is the width of the microcantilever.

For the silicon microcantilever having the dimensions $180 \times 40 \times 15$ μm and the Young modulus $E = 82$ GPa used in this work the sensitivity of the system amounted to -1.7×10^{12} Hz g^{-1}. Naturally, the microcantilever size reduction will provide an increase in the sensitivity. As follows from Eqn (3), it will be maximal in the case of reducing the microcantilever length. In turn, as follows from Eqn (2), the mass detection threshold δm is determined not only by the microcantilever dimensions, but also by the frequency measurement error δf. On the one hand, the reduction of microcantilever dimensions will lead to the growth of the sensitivity and the frequency of the eigenoscillations (and, therefore, decrease of the detection threshold). On the other hand, with the growth of the frequency one can expect an increase in the absolute error of the oscillation frequency measurement. Assuming that with the change in the microcantilever size the relative error of the oscillation frequency measurement $\delta f/f$ does not change, it is possible to show that when all three dimensions of the microcantilever are reduced by two orders of magnitude, the sensitivity increases by 10^8 times, and the detection threshold decreases by 10^6 times.

In the present study the practical realisability of reducing the mass detection threshold by using the oscillators of extremely small size was checked. For this goal a microrod produced using the method of thermal pulling at the end of a silica optical fibre served as an oscillator in the measuring system. The microrod had the shape of a cylinder with the length 15 μm and the diameter 0.3 μm (which corresponds to decreasing the length, width and thickness of the original microcantilever by 1.27×10, 1.37×10^2, and 0.57×10^2 times, respectively).

The silica optical fibre that served a base for the microrod was mounted on the electrodynamic transducer, by means of which the oscillation of microrod was excited. The object beam of the interferometer with the waist radius 17 μm was incident on the side surface of the microrod and reflected back into the crystal. Meanwhile, by the aid of the translational drive the microrod was translated along its axis with respect to the object beam. Figure 6 shows the experimentally measured dependence of the recorded signal amplitude on the position of the object beam with respect to the microrod edge. From the obtained data it is seen in Fig. 6 that until the distance z from the object beam centre to the microrod edge exceeds the beam radius, the amplitude of the demodulation signal is zero. The signal appears only when the object beam hits the microrod edge. The subsequent sharp decrease in the signal (at $z = -17$ μm), apparently, corresponds to the situation when the object beam hits the place of connection between the microrod and the silica optical fibre, which leads to presumably lateral reflection of the object beam. Further increase in the signal amplitude occurs when the beam passes the connection place and begins to hit the silica optical fibre.

The obtained result experimentally confirms the possibility of using the oscillators with transverse dimensions smaller than the wavelength of the probing radiation in microweighing systems based on dynamic photorefractive holograms. It is worth noting that the microrod used in this study had a cylindrical shape and was made of low-reflecting silica fibre (no additional reflecting coating was deposited on the microrod surface). As a consequence, the power of radiation, reflected from such a microrod, was only 5 μW, which appeared to be essentially lower than the photodetector background light, caused by scattering from crystal defects and optical elements. Nevertheless the system based on the adaptive interferometer proved to be capable of detecting oscillations of a sub-micrometre object.

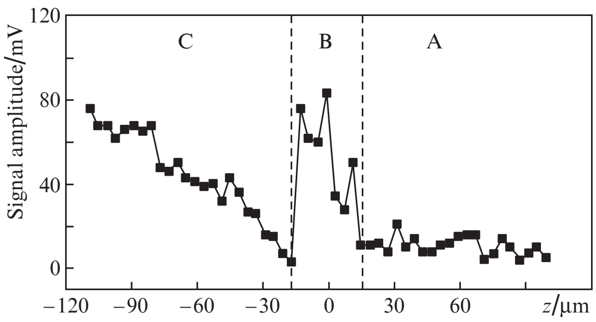


Figure 6. The amplitude of demodulated signal for different positions of the probing light beam with respect to the microrod; A – the object beam does not hit the microrod; B – the object beam hits the microrod; C – the object beam hits the base of the microrod (silica optical fibre).

Using Eqns (2) and (3) one can show that with a silicon microcantilever, comparable in dimensions with the tested microrod ($15 \times 0.3 \times 0.3$ μm), the mass detection threshold will be reduced to 5.8×10^{-19} g and the sensitivity will become as large as 2.6×10^{19} Hz g^{-1}. Thus, the reduction of the mass detection threshold by 1.5×10^7 times and the increase in sensitivity by 6.5×10^6 times is possible.

5. Conclusions

In the present study a system for measuring ultra-small masses based on resonance microweighing with the use of microcantilevers and dynamic holograms formed in a photorefractive crystal was constructed and experimentally approved. In the process of testing the measuring system the mass change of 420×10^{-12} g was detected, the mass detection threshold amounted to 8.5×10^{-12} g and the sensitivity 1.7×10^{12} Hz g^{-1}. Thanks to the adaptive properties of dynamic holograms, the system can operate in real conditions, characterised by uncontrollable variations of the environment parameters. Preliminary studies of the possibility to construct a measuring system using oscillators of submicron dimensions are carried out. Due to considerable reduction of dimensions, the use of such oscillators offers a possibility of creating systems with the detection threshold up to 5.8×10^{-19} g.

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