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論文内容要旨

Magnetic resonance imaging (MRI), as a non-invasive 3D imaging of inside structures of objects, is a well developed and widely used instrument in biology and medicine fields for human disease detection. However, owing to the sensitivity limitations of conventional inductive techniques, the 3D imaging spatial resolution of MRI is limited to several tens to hundreds of micrometer scales. With the emergency of scanning probe microscopy (SPM) techniques including scanning tunneling microscope (STM), atomic force microscope (AFM), and so on, the information of material surface can be obtained with ultra-high precision through detecting the local interaction between the sample and a probe. Based on this ultra-high force sensitivity of the probe, which is demonstrated in AFM, the resonant magnetic force from nuclear or electronic spins can be detected by a micro-fabricated sensitive cantilever. The concept of magnetic resonance force microscopy (MRFM), as a promising MRI combined with AFM technology, was firstly proposed by Sidles in 1991. This technique utilizes a small magnetic tip and an ultra-sensitive cantilever to detect the densities of spins or radicals through a non-invasive method in a nanometer scale.

Within several decades, great efforts have been performed to further improve the detection theory and exciting progresses have been achieved to further demonstrate appealing applications related to electron or nuclear spins. The experimental demonstration of MRFM was firstly reported by Rugar et al. in 1992. The first MRFM image of 1, 1-Diphenyl-2-picrylhydrazyl radical (DPPH) samples was obtained in 1993. One-dimensional nuclear magnetic resonance (NMR) imaging of semiconductor GaAs with 170 nm slice separation was demonstrated in 2003. The detection of an individual electron spin was realized with a spatial resolution of 25 nm in one dimension in 2004. Simultaneously, the 3D images of two DPPH particles were reconstructed with a spatial resolution of ~1 μm in 2004. In 2009, 3D image of tobacco mosaic viruses with resolution down to ~4 nm was detected by IBM research group. More recently in 2015, a 210 nm-wide and 32 μm-long silicon nanowire probe with a silicon mirror using a silicon-on-insulator wafer was fabricated with atto-newton sensitivity for MRFM force detection by our laboratory fellow, and scanning measurement for 3D imaging of radicals based on ESR was demonstrated. Until now, MRFM has been applied to various fields including physics, chemistry, biology and material science as an effective characterization method, with many improved aspects like using arharmonic modulation to eliminate the measurement noise, developing an ultra-sensitive cantilever for force detecting, and so on. However, this thesis is mainly focused on other two aspects of MRFM to further improve the performances of miniature MRFM system.

Firstly, back to the detection principle of MRFM, this technique uses an ultra-sensitive cantilever to detect the interacting force. But, we should know that the resolution of a cantilevered force sensor is limited by the detectable minimum force originated in thermomechanical noise. To further improve the detectable minimum force of MRFM system, usually, the measurements should be performed at low temperatures. Therefore, microscanners in MRFM for cryogenic measurements require large stroke at low temperatures with small affections to thermal variation. The performance of microscanners should be considered for the application of MRFM at cryostat measurement. Piezoelectric actuators are widely used at low temperatures; however, the full range displacement of piezoelectric actuators decreases from ~40 μm at 300 K down to ~12 μm at 170 K and ~3 μm at 1.8 K. The displacements are not enough for 3D imaging of biological species like cells. Thermoelectric actuators are driven by thermal stress, but the actuator elements are heated up due to Joule heating; therefore, its use at cryogenic applications is not basically suitable. Electromagnetic actuators cannot be applied to the MRFM system due to the leakage of magnetic field into the measurement system. In terms of magnetic force measurements for MRFM, electrostatic comb-drive actuation would be the most applicable method due to its high flexibility for system integration and relatively large actuation displacement at low temperatures. Based on conventional planar microfabrication technologies, electrostatically driven stages with large in-plane displacements can be obtained, but it is difficult to obtain large out-of-plane motion. To construct the electrostatic 3D XYZ-microstage with ability of producing large motions into XYZ directions, an independent motion mechanism to generate out-of-plane actuation force should be added.

Recently, a force-conversion mechanism with leaf springs inclined to the substrate has been utilized to transform the in-plane motion into out-of-plane motion. However, the coupled motions exist in this conversion mechanism. In order to achieve independent out-of-plane motion, the XYZ-microstage constructed with independent in-plane and out-of-plane actuators is a more attractive candidate. More recently, a 3-axis nanopositioning electrostatic microstage is fabricated with the parallel-plate structure as the out-of-plane actuation. Also, a SOI bulk micromachined XYZ-microstage is developed with comb-drive actuator in out-of-plane direction. However, the obtained out-of-plane displacements of these two electrostatic XYZ-microstages cannot meet our requirements for large 3D scanning of biological species. Normally, using monolithic wafer to realize the 3D XYZ-microstage needs to overcome the limitation of the out-of-plane motion space. Simultaneously, the out-of-plane deflection of the support springs from planer microfabrication is strongly constrained by the thickness of the device material layer in the formula of spring constant, and also the crosstalk problem happens easily due to the coupling connection between the in-plane and out-of-plane actuation units.

Microassembly technology in 3D microstructures catches burgeoning attention, because it can break through the conventional planar microfabrication processes to realize more functional devices. Here, a chip-level microassembly technology is proposed to fabricate the comb-drive XYZ-microstage, which is constructed with a comb-drive XY-microstage, two comb-drive Z-actuators and silicon based substrate. The movements of the XYZ-microstage are produced by the XY-microstage into in-plane directions and by the Z-actuators with large displacement into out-of-plane direction, respectively.

The components of comb-drive Z-actuators, comb-drive XY-microstage and silicon base substrate are designed with the detailed dimensions, respectively. To choose the suitable substrate material for comb-drive XYZ-microstage at cryostat applications, two kinds of material substrate with silicon bonded onto glass and SOI wafer are used to fabricate the comb-drive Z-actuators. The capacitive sensing also with comb-drive configuration is added in the comb-drive Z-actuator to measurement the actuation displacement. The comb-drive XY-microstage is composed of two frames: an external stationary frame and an internal movable frame. The centre plate is supported by the internal support springs connected to the internal frame and the internal frame is supported by the external support springs connected to the external frame. The centre stage can be actuated with less crosstalk into X- and Y-directions. Two kinds of support springs including serpentine spring and folded-flexure spring structures are adopted in comb-drive XY-microstages, to demonstrate that the high stiffness ratio of support springs plays an important part to achieve a large displacement.

Two kinds of comb-drive Z-actuators with the same designs are fabricated from silicon bonded onto glass and SOI wafer, respectively. The actuation performances of comb-drive Z-actuators integrated into cryostat are evaluated at room and low temperature through displacement sensor. It is demonstrated that the Z-actuator fabricated from SOI wafer possesses good actuation performance at low temperature (96 K), due to its homogenous structure to decrease the difference of expansion coefficients between two kinds of materials. The XY-microstage with folded-flexure spring as support spring can achieve large displacements, due to its large stiffness ratio of support springs. The crosstalk problem of comb-drive XY-microstage is resolved by adding some gaps for insulation in the handle layer. As the microassembly process, the comb-drive Z-actuators are vertically mounted on the grooves of silicon base substrate and kept in place by the support base block. Then, the XY-microstage is mounted onto the Z-actuators. The small pillars supported by mechanical springs in Z-actuator chips are inserted into the holes of XY-microstage. The conductive glue is used to hold the assembled structure of XYZ-microstage after curing, and also to achieve the electrical connections between the pillars in Z-actuators and the outer driving source of the XY-microstage. Therefore, all the bonded wires can be arranged together on Z-actuator chips, for the application of actuation voltages. This assembled comb-drive XYZ-microstage with a size of 12.4×15.6×16.9 mm^3 can produce large displacements of 25.2 (49.2) μm in X direction, 20.4 (27.9) μm in Y direction and 58.5 (50.5) μm Z direction. It is demonstrated that the assembled comb-drive XYZ-microstage is a promising 3D scanning stage with large displacements and less crosstalk for the application of MRFM at cryogenic environment.

To perform the MRFM experiment at room and low temperature environment, the characteristically assembled comb-drive XYZ-microstage glued with a sample of DPPH, a RF coil to excite electron spin resonant in the DPPH sample, a sensitive cantilever sensor with a small magnet to detect the magnetic resonance force, a fiber-optic interferometer to detect the vibration of the cantilever are integrated into the vacuum chamber of cryostat. At room temperature, the electron spin resonance (ESR) magnetic signals can be observed with the applied RF field from 950 to 1050 MHz when the positions of the DPPH sample are actuated to ~4 and ~12 μm displacements in Z direction, respectively. The measured magnetic resonance forces at the peaks are calculated to be 1.3×10^{-17} and 1.19×10^{-17} N, and the spin densities at the corresponding peaks are estimated to be ~1.75×10^{8} spins/μm^3 and ~1.61×10^{8} spins/μm^3, respectively. Consequently, the developed comb-drive XYZ-microstage can be applicable to MRFM measurement. Then, the vacuum chamber with a vacuum of ~8.6×10^{-4} Pa in the cryostat is cooled down using liquid N_2. However, when the temperature of the vacuum chamber is cooled down to 94.3 K, the interference signals of laser 1310 nm and 1550 nm are confused. The possible reason is that the water vapor molecular remaining in the chamber would be frozen onto the cantilever surface, which would make the stiffness of the cantilever harder. We can say that this is the equipment problem. If the vacuum system is improved, the problem can be avoided.

Normally, high vacuum environment for the resonant detection should be maintained in MRFM measurement, to decrease the air damping and increase the sensitivity of the cantilever sensor. The test objects of the sensitive cantilever are strongly constrained, because the test objects like cells should be kept in the standard atmospheric pressure environment. Therefore, vacuum packaging of the sensitive cantilever with an independent vacuum micro-chamber is a promising method to separate the vacuum environment between the cantilever sensor and test objects. Here, the second aspect in this thesis is focused on developing a vacuum packaged cantilever magnetic sensor, to construct the measurement setup of MRFM and also to realize the MRFM measurement at standard atmospheric environment.

Wafer-level bonding technology, including anodic bonding, eutectic bonding, thermocompression bonding, glass frit bonding, are very significant techniques for the devices which require to be packaged in vacuum, such as accelerometers, pressure sensors, gyroscopes and infrared microbolometer. Considering the possible influences to the bonding wafers which have already contained the fabricated small structures during the bonding process, two kinds of bonding methods including Au-Au thermocompression bonding and anodic bonding are used to realize the vacuum packaged cantilever magnetic sensors. Based on microfabrication technology, two kinds of vacuum packaged cantilever magnetic sensors are fabricated, respectively. The relationship of pressure versus quality (Q) factor of the cantilever is calibrated by laser Doppler vibrometer before the bonding process. By contrasting the measured dependence of Q factor with pressure, the packaged vacuum of cantilever magnetic sensors based on Au-Au thermocompression bonding and anodic bonding are determined to be in the range of $1.7\times10^3 \sim 2.4\times10^3$ Pa and $7.3\times10^2 \sim 1.0\times10^3$ Pa, and the force sensitivity of the cantilever sensors are estimated to be 1.4×10^{-13} N/√Hz and 1.1×10^{-13} N/√Hz, respectively.

To demonstrate the sensitivity of the vacuum packaged cantilevers to magnetic field intensity, a permanent magnet with non-uniform magnetic field is placed near the magnetized particle mounted on the cantilever tip. When the generated magnetic force is acted on the cantilever tip through the magnetic particle, the resonant frequency of cantilever will be varied, due to the changing of the effective spring constant. The magnetic field gradient can be mapped by the changing of resonant frequency. It is demonstrated that the vacuum packaging magnetic cantilever sensor based on anodic bonding are sensitive in the range from ~ 0 to ~ 15.4 mT/mm and the precision of the magnetic field measurement can reach to ~ 4×10^{-6} T.

The MRFM measurement setup is established at atmospheric environment with the vacuum packaged cantilever magnetic sensor based on anodic bonding, ~1×1 mm^2 DPPH radical, a RF coil for spin resonance and a coil for magnetic field modulation with SmCo magnet to enhance the anharmonic modulation. The resonant amplitude of the cantilever is varied with the applied magnetic field of modulation coil. The precision of the magnetic field measurement can reach to ~2.4×10^{-10} T. The ESR magnetic resonance signals are observed with the applied RF field from 500 to 1000 MHz. The measured amplitude at the peak is 7.6 nm and the corresponding force is calculated to be 9.2×10^{-12} N. The spin density at the peak is estimated to be ~1.5×10^{15} spins/cm^3.

Finally, two aspects of MRFM measurement are achieved in this thesis, to improve the sensitivity of cantilever sensor and to enlarge the measurement objects. It is demonstrated that the developed comb-drive XYZ-microstage is a promising 3D scanning stage with large displacements and less crosstalk for the application of MRFM at cryogenic environment and the developed vacuum packaging technology is an effective method to improve the sensitivity of the cantilever magnetic sensor and gives more possible applications of MRFM at various ambient conditions.

論文審査結果の要旨

試料の内部構造を非侵襲で3次元観察できる磁気共鳴イメージング（MRI）装置は，生物学や医療分野において広く使われる技術であるが，通常のMRIでは，その空間分解能は数百ミクロン程度であり，より高い空間分解能を実現するためには，試料内部により大きな磁場勾配が必要，かつ試料内部の局所的なより小さな磁化変化を検出する必要がある。走査型プローブ顕微鏡に利用されるプローブを用いて，核や電子スピンを，磁気力を用いてナノスケールの空間分解能で検出することができる磁気共鳴力顕微鏡（MRFM）が知られている。しかしながら，一般的にMRFMではノイズを低減するために極低温での計測が必要であったり，空気によるダンピングを低減するために真空中での測定が必要であったりするため，物理や化学，生物学等の分野への応用が限られる。また，低温下では通常用いられている圧電ステージの変位が小さくなるという問題がある。半導体微細加工技術をベースにしたマイクロ・ナノマシニング技術を用いると，機械要素を小型化して高感度なセンサが実現できる。近年，振動子の最小加工寸法をナノスケールまで小さくし，より高感度な力センサを実現する研究が進められている。本研究においては，微小試料を高感度でかつ高い空間分解能でイメージングするための小型のMRFMを開発することを目的とし，試料を3次元走査するために大きなストロークを持つXYZマイクロステージ，および環境の影響を受けない真空封止型の振動子（MRFMセンサ）を開発している。本論文は，これらの研究成果をまとめたものであり，全編6章からなる。

第1章は序論であり，本研究の背景，目的および構成について述べている。

第2章では，大変位が可能な静電駆動型XYZマイクロステージの設計と動作原理について述べている。長作動距離で動作する櫛歯型静電アクチュエータを利用し，1枚のXYマイクロステージと2枚のZマイクロステージをマイクロアセンブリ技術により組み立てることで，これまで実現しえなかった3軸方向の長距離動作する静電型マイクロステージを実現する手法を提案したことは重要な成果である。

第3章では，組立型の静電駆動型XYZマイクロステージを実現するための作製方法と評価結果について述べている。その性能評価結果から，マイクロステージを支持する駆動方向とその面内鉛直方向のばねの硬さ比を大きくすることでXYZ軸間の駆動クロストークの課題を改善できることを示している。これらの結果は，開発したマイクロステージが極低温でのMRFMに応用できることを示す有用な成果である。

第4章では，静電駆動型XYZマイクロステージを用いて高分子標準試料（DPPH）のMRFMによる計測を実証している。DPPH内部のラジカルを電子スピン共鳴(ESR)による磁気力変化により生じたカンチレバーセンサの振動から室温で検出することに成功している。これは，開発した静電駆動型XYZマイクロステージがMRFMに利用できることを示す重要な知見である。

第5章では，微小磁石付振動子を真空封止した小型の真空封止型MRFMセンサを開発し，大気中においてESR計測ができることを実証している。Au-Auの熱圧着接合，あるいは陽極接合を用いて微小磁石付振動子を真空封止し，その振動のQ値から内部の圧力を見積もっている。また，大気中にあるDPPHのESR計測やパーマロイ薄膜の強磁性磁気共鳴計測が可能であることを実証している。これは，微小磁石付センサを真空封止することで，大気雰囲気下でMRFM計測が可能であることを示す有用な成果である。

第6章は結論である。

以上要するに本論文は，新たに開発した長作動距離で動作する静電駆動型XYZマイクロステージ，および真空封止型のMRFMセンサを開発し，小型磁気共鳴力顕微鏡システムを実現したものであり，これらの成果は，機械システムデザイン工学およびナノ機械工学の発展に寄与するところが少なくない。

よって，本論文は博士(工学)の学位論文として合格と認める。