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Dissertation Thesis Abstract

Study of materials magnetic properties for direct radiation energy conversion into electricity

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Abstrakt

Nízka konverzná účinnosť termoelektrických materiálov využívaných v rádioizotopových termálnych generátoroch a pomalé napredovanie napriek nemalému výskumnému úsiliu motivuje snahy o hľadanie nových princípov, ktoré by poskytli vyššiu konverznú účinnosť.

Cieľom tejto práce bolo skúmať termomagnetickú konverziu energie ionizujúceho žiarenia. Termomagnetické generátory dnes väčšinou pozostávajú s pohyblivých častí, alebo piezoelektrických strún, kvôli využívaniu zdrojov s konštantným tepelným výkonom. Štúdiom brzdenia iónov v magnetických materiáloch v súvislosti s ultrarýchlou demagnetizáciou bol navrhnutý a experimentálne študovaný nový prístup k termomagnetickej konverzii ionizujúceho žiarenia.

V rámci predkladanej práce boli najprv vykonané testy s nanosekundovým a femtosekundovým laserom, za účelom štúdia termomagnetickej konverzie využitím termomagnetického efektu a ultrarýchlej demagnetizácie. Indukčné merania potvrdili konverziu energie na nanosekundovej a sub-nanosekundovej škále a poskytli informácie o faktoroch ovplyvňujúcich konverznú účinnosť.

Simulácie tepelného efektu spôsobeného brzdením iónov a testy na urýchľovači IC100 so 150 MeV Xe iónmi a alfa časticami emitovanými Ameríciom 241 boli vykonané pre potvrdenie využiteľnosti termomagnetickej konverzie energie tepelného efektu spôsobeného brzdením jednotlivých iónov.

Abstract

The relatively low conversion efficiency of thermoelectric materials used in radioisotope thermal generators and slow progress in the field, despite continuous research efforts, motivate a quest for new conversion principles, which might offer higher conversion efficiency.

The aim of this work was to investigate thermomagnetic energy conversion of ionizing radiation. Nowadays, thermomagnetic energy conversion devices usually incorporate moving parts or piezoelectric springs, due to the constant power heat sources used. By studying the interaction of ion-stopping in magnetic materials and relation to ultrafast magnetization effects, a new approach to thermomagnetic power conversion has been proposed and experimentally studied.

First, tests with nanosecond and femtosecond lasers were carried out in order to study thermomagnetic power conversion using thermomagnetic effect and ultrafast demagnetization. Inductive measurements proved thermomagnetic power conversion on nanosecond and sub-nanosecond scale viable and provided information about the factors influencing conversion efficiency.

Simulations of the ion-induced thermal spike and tests at the accelerator IC100 with 150 MeV Xe ions and with alpha particles emitted by Americium 241 radioisotope were conducted with the aim to confirm the feasibility of thermomagnetic power conversion utilizing ion-induced thermal spike.

Goals of the thesis

1. Experimentally study the thermomagnetic effect induced by ions braking in ferromagnetic materials.

2. On the experimental results basis, assess the thermomagnetic effect achievable efficiency.

3. Propose a potential technical solution for the conversion of ionizing radiation energy by means of a thermomagnetic effect.

4. Identify possible applications of the studied conversion scheme.

1 Introduction

The area of radiation energy conversion systems is a wide field, with many different conversion technologies proposed and tested. However, the most widely used technology today, the thermoelectric conversion employed in radioisotope thermal generators (RTGs), works with efficiency just around 6%, with the best performing thermoelectric cells delivering 15% under ideal conditions [1]. This leaves space for other technologies, with potentially higher efficiency, such as thermophotovoltaic conversion [2], alpha or betavoltaics and dynamic generators, like Stirling generators [3]. All of the mentioned technologies could provide higher efficiency, however, their low radiation hardness, or low reliability, due to moving parts and other factors, makes the thermoelectric conversion the best choice for most space applications using radioisotope power sources nowadays.

Therefore, the quest for new conversion principles enabling higher efficiency or being more suitable for specific applications in space or on the ground is still on-going. With the calculated theoretical thermomagnetic efficiency being on the order of 55% of Carnot, it is reasonable to ask whether a thermomagnetic power conversion could not provide higher conversion efficiency than betavoltaics or any other current technology. Although experimental values indicate efficiency lower than 25%, new materials employing order-to-order phase transitions were shown to have potential for achieving higher efficiencies. The efficiency of 30% relative to Carnot is expected for a single domain Gd [4].

However, for a thermomagnetic generator either an impulse heat source is required, or the generator must employ moving parts. The advantage of using radioisotope power source lies in the fact that it is able to provide an impulse heating, when considering single ion heating on a microscale, thus eliminating the need for moving parts. The ion-induced thermal spike model describes heating in the vicinity of the particle track due to ion stopping. Based on the model, and confirmed by experiments, temperatures high enough to locally melt metals were observed after swift heavy ion impact [5].

Furthermore, the heating in the particle track happens on a femtosecond time scale, suggesting ultrafast demagnetization (UFD) processes might be involved [6], which could improve overall efficiency of the conversion if energy could be extracted from the electron and spin system, before the heat is dissipated to the lattice.

Purpose of this work is to examine the possibility of using magnetic properties of materials, associated with the thermomagnetic effect and ultrafast demagnetization, for radiation power conversion and to propose and conduct experiments able to answer the question of the feasibility of power conversion scheme based on the ion-induced thermal spike and to assess achievable efficiency.

2 Overview of the current status

2.1 Ultrafast demagnetization

2.1.1 Ultrafast laser induced demagnetization

When ultrafast laser-induced demagnetization was first reported, the authors interpreted their result in terms of three temperature model (3TM). This model describes energy flow between three different systems, which are assumed to be in internal equilibrium: electron, lattice, and the spin system. An electron temperature T_e, a lattice temperature T_p and a spin temperature T_s are defined for the three systems as it can be seen in figure 1 [7]. The electron system is heated by the optical excitation almost instantaneously and its energy is rapidly transferred to the spin system causing the initial rapid demagnetization on the order of hundreds of femtoseconds. The subsequent thermalization of the electron and spin system with the lattice after several picoseconds results in a partial recovery of the magnetization. On a timescale of a few nanoseconds, the three subsystems cool down because of heat transfer to the surrounding material, and the magnetization recovers completely [8].

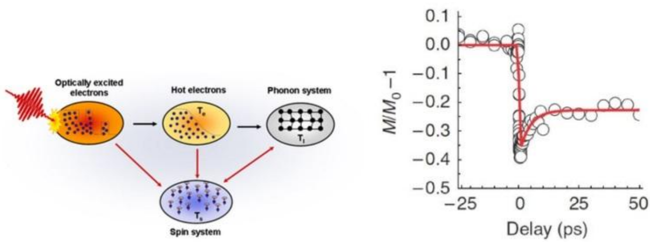

Fig. 1 A graphical representation of the three temperature model, where T_e is electron temperature, T_s spin system temperature and T_l lattice temperature and an actual demagnetization characteristic of a CoPd multilayer film [8].

3 Particle stopping induced thermomagnetic effect

The recent confirmation of indirectly excited ultrafast magnetization dynamics opens up a possibility to initiate UFD and related magnetization precession also by other than optical sources of excitation.

On the first glance, it seems that no conventional heat source suitable for power generation fulfils this condition. However, with a closer look at radioisotope, or nuclear power sources, one finds that the radiation heating employed in nuclear reactors or RTG's is not just simple continuous heating. The observed continuous heating is a summary effect of fast heat pulses, so-called thermal spikes, created during individual nuclear reactions and subsequent particle stopping. In a small volume around the high energy particle track, the heating can be considered impulse. Simulations of ion-induced thermal spike and experiments done in this field confirm very fast, even sub-femtosecond heating of the track area [9]. Simulations of the thermal spike in a particle track of alpha particles and swift heavy ions in water have yielded a temperature increase from 400K up to 10^4K respectively along the particle track axis, with the time constant of the temperature drop on the order of 10^{-11} s [9].

By comparing the temperature calculated using the thermal-spike model at a given time and radius around the track with the Curie temperature, a rough estimate of the demagnetized radius could be obtained as shown in figure 2.

The heating throughout and near the particle track during ion stopping is a long-known effect. However, it has not yet been considered for power conversion, using the thermomagnetic effect to the best of the author's knowledge. To assess the feasibility of the power conversion, it is necessary to combine both experimental and theoretical knowledge of ultrafast magnetization and ion-induced thermal spike.

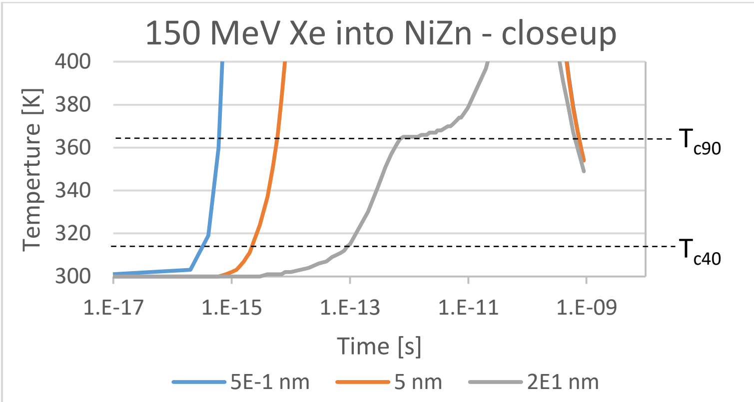

Fig. 2 Thermal spike simulation of a 150 MeV Xe ion in NiZn ferrite with lines representing curie temperatures of 40 (T_{c40}) and 90°C (T_{c90}) is shown in the figure for 0.5, 5 and 20 nm radius around the particle track.

Due to complicated simulation required for accurate assessment of the induced voltage using a magnetically soft sample and to prove the actual samples are able to convert fast heat pulses into electrical energy, experiments with laser-induced demagnetization were conducted. Those served as a benchmark for amplitude assessment of the ion-induced demagnetization.

4 Experiments with thermomagnetic conversion of laser pulses

4.1 The measurement setup

Lasers with pulse widths on the nanosecond and femtosecond time scale were used to induce fast change of magnetization in samples. The setup consisted of a CryLas laser, (amplifiers), silicon PIN diode detector and a LeCroy WaveMaster 808Zi oscilloscope with sampling rate of 40 GS/s.

The signal from the semiconductor diode detector has been used as a trigger signal. The block diagram of the measurement circuit is shown in the figure 3.

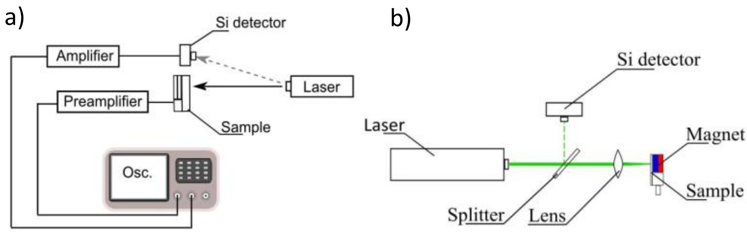

Fig. 3 Block scheme of the measurement setup (a) and the optical setup layout (b)

The same principle of triggering by Si detector and averaging of the signal was used also in measurements with a femtosecond laser Spectra Physics Spirit 4C. The femtosecond laser had a pulse width of 220 fs and adjustable pulse energy up to 40 µJ. Due to higher energy per pulse, amplification was not required, lowering signal distortion by electronics to minimum.

4.2 Samples and tested materials

Samples of magnetically soft material were deposited onto an inductor, prepared on a printed circuit board by means of photolithography. The inductor terminals were connected to an SMA connector, and a demountable permanent magnet was attached to the other side of the PCB. The loop antennas showed best results in terms of signal strength and amplitude. The loop antennas are shown in the figure 4.

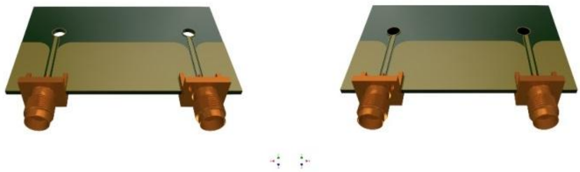

Fig. 4 3D view of the second generation of loop antenna designs, where a design of a 1mm diameter loop antenna with a hole (right) and without hole (left) is shown.

The return loss parameter was measured for the new designs in the range of 10 kHz to 40 GHz.

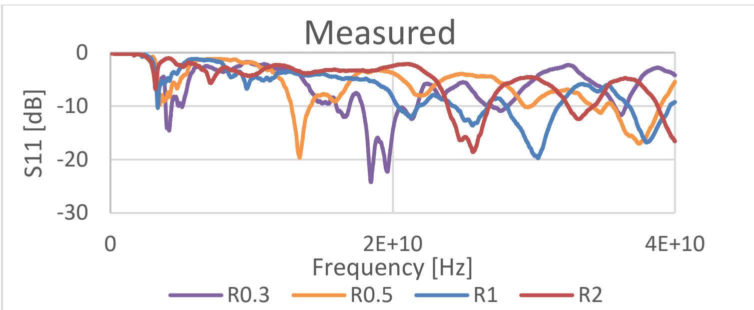

Fig. 5 Measurement of the return loss parameter for loop inductor designs. (Credit: Asoc. Prof. Martin Tomáška, PhD.)

Depositions of $NiZnFe_2O_4$ layers were carried out in argon atmosphere by a high voltage DC power source and a magnetron sputtering device. Two compositions, namely $Ni_{0.33}Zn_{0.67}Fe_2O_4$ and $Ni_{0.3}Zn_{0.7}Fe_2O_4$ with Curie temperatures of 90 and 40°C respectively, were used in experiments.

After first successful tests of the measurement scheme, more magnetic materials were tested with the femtosecond laser. Among them a 40 µm commercial Gadolinium foil prepared with three different coatings: Gold, SiO_2 and Diamond-like-carbon, all with 50 nm thickness. Later, also a 50 nm Cobalt layer was tested.

4.3 Femtosecond laser measurements

Similarly to the experiments with nanosecond laser, a test whether the observed effect is of a thermomagnetic origin was conducted by comparing characteristics measured under two opposite field polarities and a reference measurement without applied magnetic field as it can be seen in the figure 6.

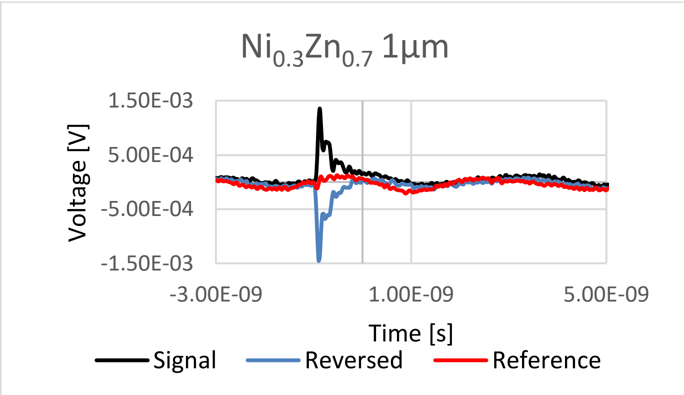

Fig. 6 Characteristics measured using a 1 micron thick sputtered $Ni_{0.3}Zn_{0.7}Fe_2O_4$ layer with the loop diameter of 2 mm under applied external magnetic field (signal), without external magnetic field (reference) and with the external field direction reversed (reversed).

The observed signal raise time and signal duration were confirmed to be dependent on the thickness of the ferromagnetic layer. Figure 7 shows a comparison of rise times observed for $Ni_{0.33}Zn_{0.67}Fe_2O_4$ samples at several different thicknesses.

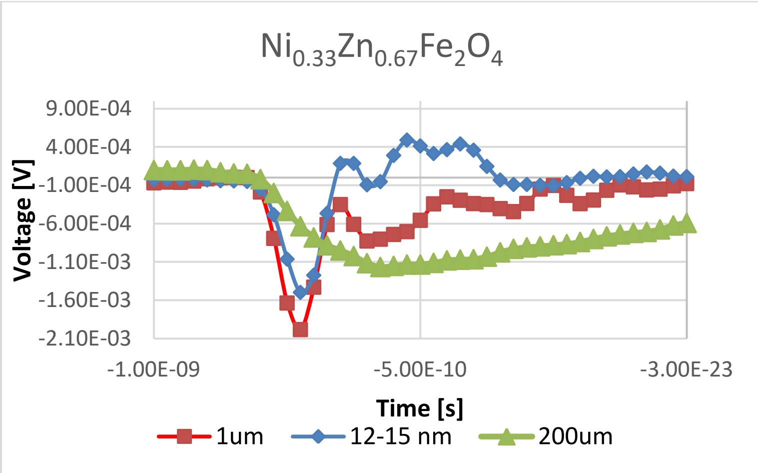

Fig. 7 A comparison of rise times of demagnetization induced signals from $Ni_{0.33}Zn_{0.67}Fe_2O_4$ powder sample with 200 µm grain size, 1 µm and 12-15 nm sputtered layer using a 1 mm diameter inductor.

Frequency range of the used measurement equipment limited the fastest observable signal rise time to 75 ps. All sputtered layers, both $Ni_{0.33}Zn0._{67}Fe_2O_4$ and $Ni_{0.3}Zn_{0.7}Fe_2O_4$, with thickness below 1 um and also Fe-Ni alloys showed rise time of 75 ps, while thicker samples showed longer rise-time.

The role of heat transfer was confirmed by a signal from the 40 µm thick Gd foil, coated by a 50 nm Au, SiO_2, and Diamond-like Carbon (DLC). Measurements of induced voltage from a 50 nm Cobalt layer further supported the role of heat transfer, as a re-magnetization peak was clearly visible for Co, due to its Curie temperature on the order of 1000°C. Due to permanent magnetization of cobalt; it was not necessary to apply external magnetic field; since only a part of the layer reached the Curie temperature. However, a clear response to the external magnetic field was observed, when applied.

5 Alpha particle induced thermomagnetic effect experiments

The measurement setup for alpha particle induced demagnetization experiments, was based on the same principle of triggering and amplification as the scheme for laser induced demagnetization tests

The scintillation triggering was provided by a fast timing plastic scintillator, processed into a 2x2x20 mm block attached directly to a Sensl MicroFJ-SMA-30035 silicon photomultiplier (SiPM) with a fast output and rise time below 100 ps. ORTEC VT120B amplifier was used to amplify SiPM signal to be used as trigger. Several amplifiers, including Minicircuits ZKL-1R5+ with 40 dB amplification up to 1.5 GHz, were used to amplify the signal. The silicon photomultiplier and sample were placed in a faraday cage to minimize background noise.

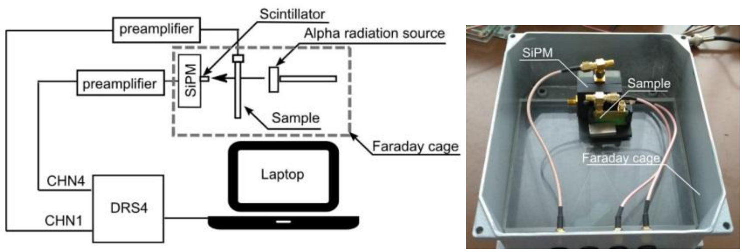

Fig. 8 A schematics (left) and actual (right) layout of the experimental set-up, used in alpha particle induced thermomagnetic effect experiments.

DRS4 was used as an oscilloscope to provide averaging of more than 3000 signal characteristics.

After successful laser tests with Gadolinium with low Curie temperature and high thermomagnetic efficiency, plasma etching and pressing was used to decrease thickness of the commercial 25 µm thick Gd foil below 10 µm, to allow 150 MeV Xe ions and

5.5 MeV alpha particles to pass through. After processing, a 50 nm protective DLC coating was implemented.

5.1 Results and discussion

The initial measurements using NiZn ferrite deposited on the PEN foil, were conducted to test the triggering system and interference. Triggering by alpha particles passing through the sample was demonstrated by comparing the amount of signal counts per second with the radioisotope and without it. After removing alpha radioisotope from the vicinity of the sample, the amount of trigger signals decreased to zero.

Signal averaging reduced apparent noise level down to approx. 6 µV peak-to-peak as it can be seen in the figure 9, already accounting for used 40 dB (100 times amplitude) amplification. Further decrease of the noise level by subtraction of measurements with different field polarities resulted in noise level below 4 µV pk-pk on average.

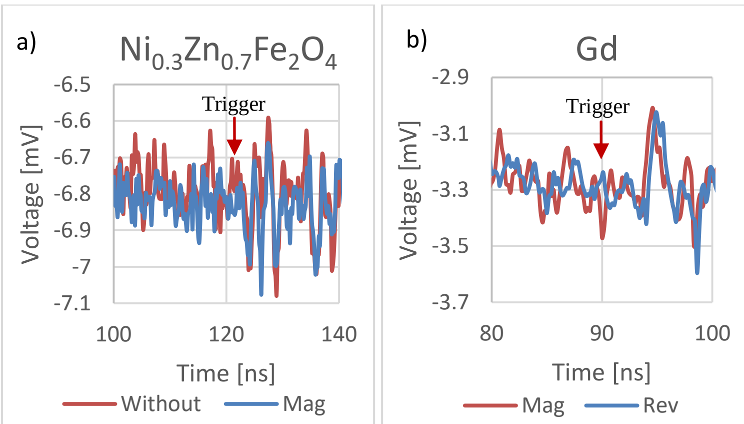

Fig. 9 Measured and averaged characteristic of 1 µm $Ni_{0.3}Zn_{0.7}Fe_2O_4$ ferrite deposited on 12 µm PEN without applied magnetic field (red) and with applied field (blue) (a) and a plasma etched Gadolinium membrane with applied field (red) and field direction reversed (blue) (b).

A small interference was observed in all measurements, however, subtraction of the signals measured under different magnetic field polarity allowed the interference pattern to be removed.

High timing precision and a known delay between measurement and triggering circuit, allowed the position of the signal on the time axis to be localized precisely.

However, despite 0.2 ns timing precision, no signal was observed in the alpha particle, nor in measurement with 150 MeV ions at the accelerator, suggesting signal amplitude to be less than 5 µV. Considering 15 µV amplitude, 0.1 ns rise time and 2 ns exponential decay to half[1] the energy of the demagnetization pulse was lower than 0.0624 eV - near thermal noise. Such amplitudes are too low for most applications, not to mention energy conversion.

6 Feasibility and efficiency assessment of thermomagnetic conversion for fast impulse heat sources

6.1 Theoretically achievable efficiency

First estimates regarding conversion efficiency were made based on the theoretical thermomagnetic efficiency of the common magnetocaloric materials, such as Iron, Nickel and Gadolinium. The theoretical efficiencies for these materials are on the order of 3 – 12% relative to Carnot efficiency [10]. However, calculations for single domain Gadolinium and $NdCo_5$ exhibiting spin reorientation show efficiencies on the order of 30 – 45% relative to Carnot efficiency. Furthermore, the ultrafast demagnetization, confirmed to be connected with ion-stopping [6], might allow higher

[1] Calcualted using Vishay pulse energy calculator

thermodynamic efficiency, as there are three heat reservoirs involved.

There are however practical problems, besides materials engineering, to achieve the theoretical efficiencies. The thermodynamic efficiency of the static thermomagnetic conversion cells/samples, used for power conversion in our experiments is lower than in a conventional thermomagnetic cycle, which is a trade-off for reliability and inevitable for the used sub-nanosecond heat pulse duration associated with expected GHz - THz output frequencies.

6.2 Experimentally achieved efficiency

The experimental efficiencies calculated from voltage signals acquired in femtosecond laser tests, converted to energy using Vishay online pulse energy calculator for $Ni_{0.33}Zn_{0.67}Fe_2O_4$, Gadolinium and Cobalt samples are listed in the Tab.1.

Tab. 1 Total signal energy estimates calculated for signals from 200 µm $Ni_{0.33}Zn_{0.67}Fe_2O_4$, Gadolinium with 50 nm Au layer and a 50 nm Co layer, vs laser input pulse energy and total efficiency.

Material	Impulse parameters				Values		
	trise [ns]	td1/2 [ns]	Vpeak [mV]	R [Ω]	Epulse [aJ]	Elaser [µJ]	η [%]
NiZn	0.2	1	1.2	50	18.5	40	4.64E-11
Co	0.1	0.2	6	50	75.9	40	1.90E-10
Gd	0.1	10	1	50	143	40	3.59E-10

The efficiencies calculated using exponentially decaying pulse approximation are very low, therefore without substantial increase in efficiency, a power conversion using the proposed conversion cells is not viable.

Assuming the same efficiency for ion-induced demagnetization signal, pulse energies for 5.5 MeV alpha particle and 150 MeV Xe ion, considering whole energy deposited in the Gadolinium layer, would be 3.16×10^{-22} J (0.002 eV) and 8.62×10^{-21} J (0.054 eV) respectively – on the edge of detection limits of the designed detection scheme for Xe ions and a little below the detection limits of the detection scheme for alpha particles.

The low efficiency of the conversion cells is believed to be associated with wide bandwidth of the signal and high return loss at frequencies, where maximum of the signal energy is concentrated according to FFT, and due to low thermodynamic efficiency (associated with static thermomagnetic cycle and low Carnot efficiency for low temperature difference) at room temperature, where the thermomagnetic efficiency was shown to be highest according to estimates based on the thermal spike model.

7 Conclusion

In this thesis, a new mechanism for alpha radiation power conversion based on the ion-induced thermomagnetic effect was proposed. The expected existence of the ion-induced thermomagnetic effect is justified by the thermal spike model and experiments proving ion-induced demagnetization on a nanometer and femtosecond scale.

The thermomagnetic power conversion of fast, femtosecond laser-induced heat pulses by magnetic loop antennas was proved in the experimental part. Several conversion cell designs employing NiZn ferrite, Gadolinium, Cobalt, and Fe_1Ni_3 alloy magnetic layers were prepared and tested.

Experimental setup for measurement of ion-induced and alpha-particle induced demagnetization was developed, having output signal detection limits below 0.054 eV at 1 GHz.

However, measurements with ions and alpha particles did not show any result within the detection limits. The deeper analysis uncovered several reasons for unexpectedly low efficiency. One of them was a broad frequency range of the signal, too broad for most known antenna designs. This, together with a highly localized source – 1s to 100s of nm around the particle track, resulted in low field-change conversion efficiency. Another reason was low thermodynamic efficiency due to a static conversion cell design, required for high-frequency operation.

Ways to significantly improve the efficiency have to be identified, for the thermomagnetic conversion of radiation via ion-induced thermal spike to be considered for real-world applications. Nevertheless, after further development, the conversion cells could be used as fast, broadband detectors for femtosecond laser testing, or pump-probe experimental schemes, providing rise times on the order of picoseconds.

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