

Patterning of phase change films with microlens arrays

Y. Lin^{a,b,c}, M.H. Hong^{b,c,*}, G.X. Chen^a, C.S. Lim^d,
Z.B. Wang^c, L.S. Tan^b, L.P. Shi^c, T.C. Chong^{b,c}

^a Nanoscience & Nanotechnology Initiative, National University of Singapore, Singapore 117576, Singapore

^b Department of Electrical and Computer Engineering, National University of Singapore, Singapore 117576, Singapore

^c Data Storage Institute, Singapore 117608, Singapore

^d Department of Mechanical Engineering, National University of Singapore, Singapore 117576, Singapore

Received 30 September 2005; received in revised form 16 January 2006; accepted 1 February 2006

Available online 22 January 2007

Abstract

Nanometer-sized features were fabricated on Ge₁Sb₂Te₄ phase change films uniformly by irradiation with the fundamental and second harmonic components of a Nd:YAG laser through a microlens array. The effect of laser fluence and laser wavelength on the feature size was investigated. The change in the morphology and optical property of the phase change film was characterized. The smallest feature size of 355 nm was produced by using a low laser fluence and the 532 nm second harmonic of the laser. This was much smaller than the focus beam size of the microlens. The laser irradiation caused the formation of crystalline features on the amorphous phase change film. The amorphous region of the film could be removed by dipping in a NaOH solution, leaving the crystalline regions unaffected.

© 2006 Elsevier B.V. All rights reserved.

Keywords: Phase change; Microlens array; Atomic force microscopy; Near-field scanning optical microscopy

1. Introduction

Phase change materials have been very popular in optical data storage field in producing rewritable compact disks (CD-RW) and digital versatile disks random access memory (DVD-RAM) [1,2]. A phase change film is made from compound materials, whose phase is changed from amorphous to crystalline state and reversely by heat treatment. The data information can be recorded by the difference of optical reflectivities of the crystalline and amorphous states. The field of optical recording is facing the challenge of making the recording mark size as small as possible due to the limitation of focused laser beam size. The full-width-at-1/e²-irradiance level for a focal spot size can be expressed approximately as $s = \lambda / (n \sin \theta)$, where θ is the marginal ray angle and n is the refractive index of the medium at focus. The denominator ($n \sin \theta$) is defined as the numerical aperture (NA). In order to maximize disc capacity, the optical system should have either a higher NA or a shorter

λ to get a smaller focal spot size. To get a higher NA, near-field probe techniques, such as near-field optical lithography by atomic force microscopy (AFM) and near-field scanning optical microscopy (NSOM), have been developed to overcome the diffraction limit to reduce the feature size in the phase change film [3]. However, these techniques can only record over a very small area at a time and therefore have a low throughput. In this work, microlens array (MLA) is proposed to be used for patterning in a phase change film (Ge₁Sb₂Te₄, GST film). A MLA consists of many miniaturized lenses with same size and focal length in the order of microns. These lenses can translate the laser irradiation into a series of focal points as light pens to fabricate a large number of features simultaneously. The capability of MLA in parallel processing has been used for real-time imaging with laser scanning confocal microscopy [4–7], where the laser beam is converted to hundreds of small focal points at its focal plane. In recent years, MLA was also used in photolithography [8,9] and it was found that micro- and nano-features can be produced over large area repetitively in a simple manner. In this article, the fundamental and second harmonic components (at wavelengths of 1064 and 532 nm, respectively) of a Nd:YAG laser are used to irradiate a GST film through a MLA to generate crystalline features on the GST.

* Corresponding author at: Data Storage Institute, Singapore 117576, Singapore. Tel.: +65 6874 8707; fax: +65 6777 1349.

E-mail address: HONG.Minghui@dsi.a-star.edu.sg (M.H. Hong).

The effect of the incident laser fluence on feature size is investigated.

2. Experimental

Fig. 1 shows the optical image of a MLA and the schematic diagram of the experimental setup. The number of lenses, which are made of fused silica, is 401×401 in an area of $10 \text{ mm} \times 10 \text{ mm}$. The diameter of the lenses is $23 \mu\text{m}$ and the lenses are arranged in a hexagonal array with a pitch of $25 \mu\text{m}$. The sag of each lens is $9 \mu\text{m}$ in height, which is equivalent to a focal length of $28.7 \mu\text{m}$, thus giving a numerical aperture (NA) of 0.59. The GST film is 100 nm thick, coated on a polycarbonate substrate, and its original state is in the amorphous phase. The fundamental and second harmonic components of the Nd:YAG laser beam (BMI 5000, pulse duration 7 ns), at wavelengths of 1064 and 532 nm, respectively, are used to irradiate the sample through the MLA at different laser fluences. The features on the GST film are characterized by atomic force microscopy (AFM, DI 3100) and near-field scanning optical microscopy (NSOM, Aurora-2, Veeco) in the transmission mode.

3. Theory

Fig. 2 shows the atomic arrangement of the NaCl type crystals in the crystallized GeSbTe film [10,11]. At non-focal planes of the MLA, interference between lenses occurs. The light intensity distribution at planes with different distances to the MLA is quite different [12]. The foci at the focal plane of the MLA form a periodic array as the microlens arrangement, and the focal plane is set at $z=0$. The complex wave amplitude $u(x, y, z=0)$ of the

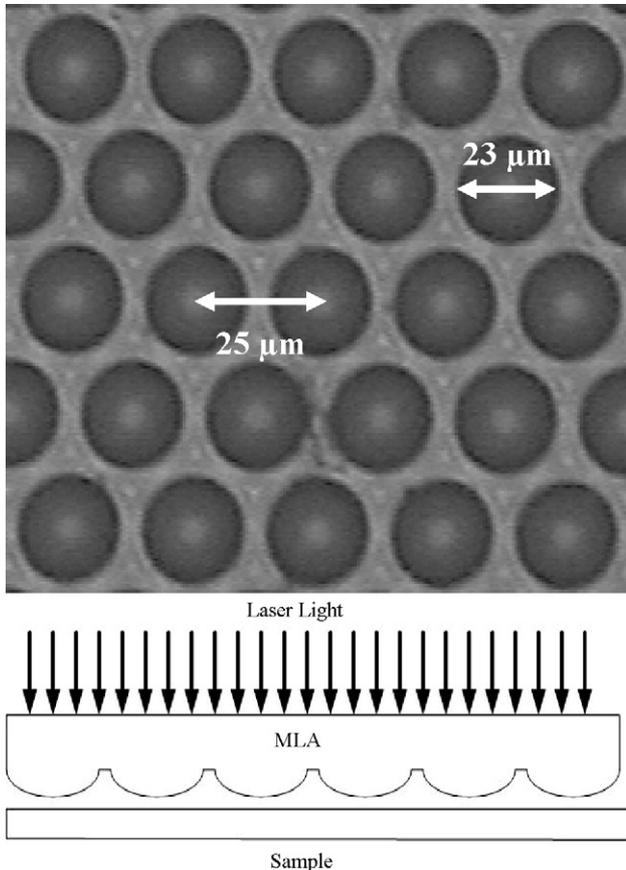


Fig. 1. Optical image of a microlens array (MLA) and schematic diagram of the experimental setup.

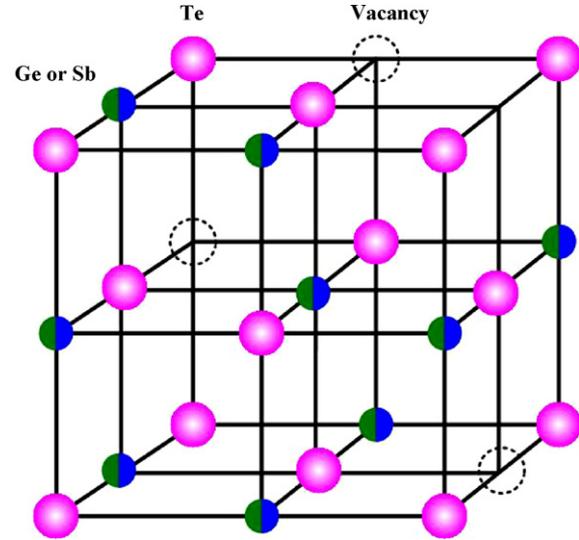


Fig. 2. Atomic arrangement of the NaCl type crystals in the crystallized GeSbTe films.

foci can be represented by the convolution of the wave amplitude $A(x, y)$ of a single lens focus and the array generating function $g(x, y)$:

$$u(x, y, z=0) = A(x, y) \otimes g(x, y). \quad (1)$$

The wave amplitude in a fractional Talbot planes can be expressed as:

$$u\left(x, y, z = \frac{M}{N}z_T\right) = \exp\left[2\pi i \frac{Mz_T}{N\lambda}\right] \sum_{k=-\infty}^{+\infty} \sum_{l=-\infty}^{+\infty} F_{k,l,M,N} A\left(x - k\frac{p}{N}, y - l\frac{p}{N}\right). \quad (2)$$

In this equation, z_T is the Talbot length (the distance where intensity distribution repeats itself immediately after the focus plane [13]), and M and N are non-negative integers, while p is the period of the microlens. F is the complex factor, which is responsible for multiple focus points at the fractional Talbot plane. The light intensity I is proportional to the square of the wave amplitude, so it is related to F and A as:

$$I(x, y, z) \propto |F|^2 \left| A\left(x - k\frac{p}{N}, y - l\frac{p}{N}\right) \right|^2. \quad (3)$$

The intensity at one single lens focus is proportional to $|A|^2$, and its peak is at the centre of the axis. If A is zero in the range outside p/N , there will be no overlap between the multiplied functions A and the intensity at each focus is same and proportional to $|F|^2$; if A is non-zero in the range outside p/N , overlap will happen and the intensity depends on both $|F|^2$ and $|A|^2$, which leads to the multiple foci in the fractional Talbot planes having different intensities. If only one Talbot length is considered, $M=1$ and $z=(1/N)z_T$. When N is large, where z is small, i.e. the fractional Talbot plane is very near to the focal plane, $|A|^2$ outside the range of p/N is large and interference effect is great.

4. Results and discussion

Fig. 3 shows the optical microscopy image of a series of dot features fabricated by laser irradiation at a fluence of 39.4 mJ/cm^2 , with the GST phase change film placed at different distances from the MLA. The crystalline features display higher reflectivity than the amorphous surroundings. Since the separation between the MLA and the GST films were only about $10 \mu\text{m}$, it was difficult to measure the exact distance, only the qualitative relationship between distance and feature arrangement was considered. When the film was placed at the focal plane, the dot features were patterned by the foci of the lenses directly and the feature size was around $1 \mu\text{m}$, as shown in Fig. 3(a). When the sample was placed away from the focal plane, multiple foci resulted in extra dot features as well as different dot feature sizes, as shown in Fig. 3(b)–(d). The largest dot features were created by the foci of the microlenses, while the smaller features were due to interference at fractional Talbot planes. As discussed above, a shorter separation distance leads to greater interference and more multiple foci, so the features shown in Fig. 3(b) were obtained at the nearest distance to the focal plane, while those in Fig. 3(d) were the furthest.

The minimum focus spot size of the lens depends on the laser wavelength, as described in Eq. (4), where M^2 denotes the laser beam profile quality, λ is the wavelength, f is the focus length, and d_0 is the size of the lens. The laser beam was approximated as a Gaussian profile, so M^2 is equal to 1. When the lens parameters are fixed, the minimum focus size increases with the wavelength linearly, i.e.:

$$d_{\min} = \frac{4M^2\lambda f}{\pi d_0}. \quad (4)$$

The fundamental and second harmonic components of the Nd:YAG lasers were used to irradiate the film through the MLA to study the wavelength effect on the pattern size. For the wavelengths of 1064 and 532 nm, the calculated minimum focus spot sizes are around 1690 and 845 nm, respectively. For each wavelength, laser fluences from the highest to lowest were tested for patterning. Since the phase change temperature of the GST film from the amorphous to crystalline state is around 200°C , too high an incident laser fluence will ablate the GST film to form holes. Fig. 4 shows the feature sizes obtained at different incident laser fluences for 532 nm laser

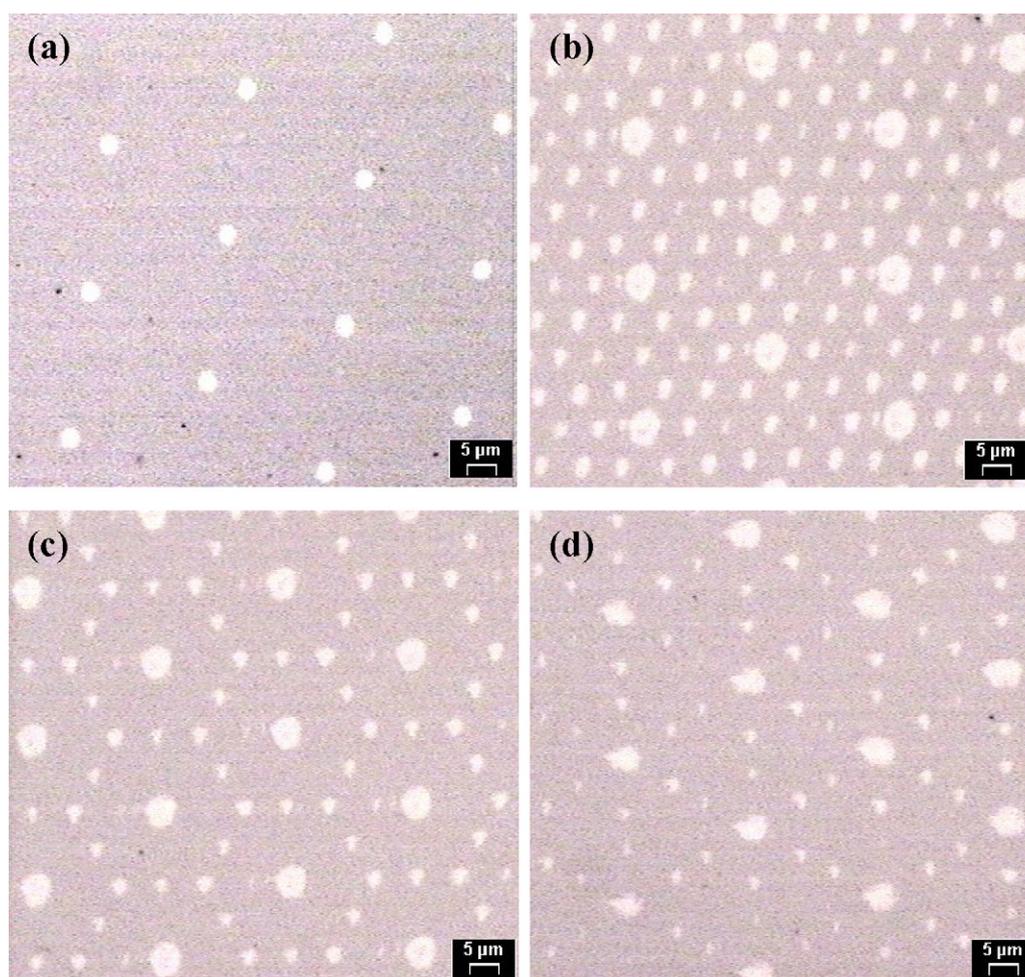


Fig. 3. A series of optical micropatterns produced in phase change film by the MLA with the same incident laser fluence at 39.4 mJ/cm^2 but different distances between the MLA and phase change film.

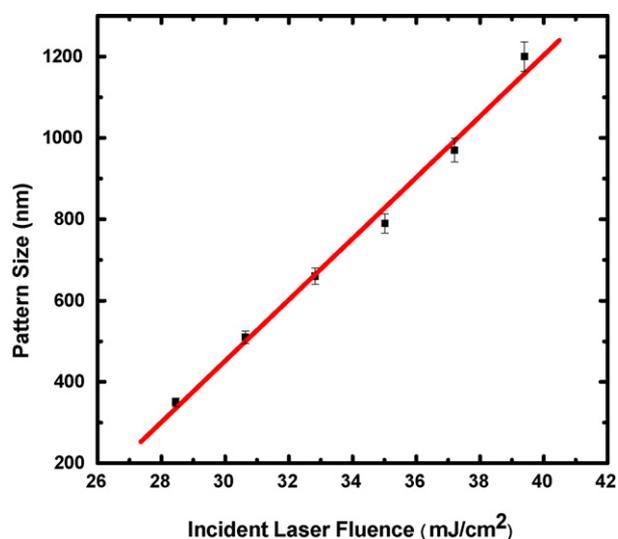


Fig. 4. Dependence of feature size on incident laser fluences with 532 nm of Nd:YAG laser irradiation.

irradiation. It shows that the pattern size increases with laser power linearly, which was also observed for 1064 nm laser irradiation.

The patterned GST film was characterized by AFM and NSOM in the transmission mode. The optical transmissivity of the film in the crystalline state is lower than that in the amorphous state [14], as shown in Fig. 5. The dot features were fabricated by laser irradiation at 532 nm with incident laser fluences of (a) 37.2 mJ/cm² and (b) 28.4 mJ/cm², and the sizes are around 800 and 355 nm, respectively. For Fig. 5(a), the AFM image shows that the morphology of the dot feature, other than the phase state, has changed, and a bump was formed. It means that the incident laser fluence of 37.2 mJ/cm² is around the critical laser ablation energy of the phase change film. When the laser fluence is decreased, no morphology change is detected. At a laser fluence of 28.4 mJ/cm², the smallest feature size of 355 nm for laser irradiation at 532 nm wavelength is obtained. The smallest feature size formed by laser irradiation at 1064 nm is around 512 nm. It is noticed that the minimum feature sizes obtained are much smaller than the focus beam size of the MLA calculated using Eq. (1). This indicates that the limitation of focus size can be overcome by using a lower laser fluence. The laser pulse can be approximated as a Gaussian shape. When the incident laser fluence is low, only the peak power of the laser pulse is able to heat the film to phase change temperature, which makes the affected area smaller than the focus size of the lens.

The patterned GST film was dipped into 30% NaOH solution for 2 min to study the different reaction of amorphous and crystalline phase states to an alkaline solution. Fig. 6 displays the three-dimensional AFM image of the patterned phase change film after it was dipped in the NaOH solution. The height of each feature is 100 nm, which is exactly equal to the film thickness. The NaOH solution dissolved the amorphous region of the film while the crystalline dots were not affected, which resulted in the formation of dots in pillar shape. Although the mechanism

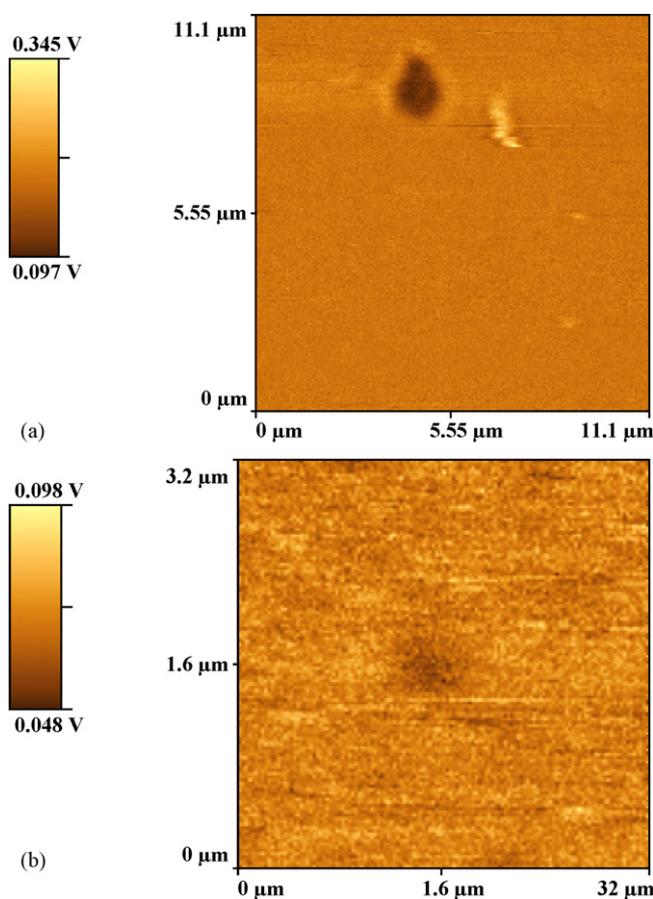


Fig. 5. NSOM transmission images of patterns in the phase change film irradiated by 532 nm Nd:YAG laser fluence of (a) 37.2 mJ/cm² and (b) 28.4 mJ/cm².

of this chemical phenomenon is still not clear yet, this method can be used to produce three-dimensional structures in the phase change film and thus provides a new material processing method for phase change films.

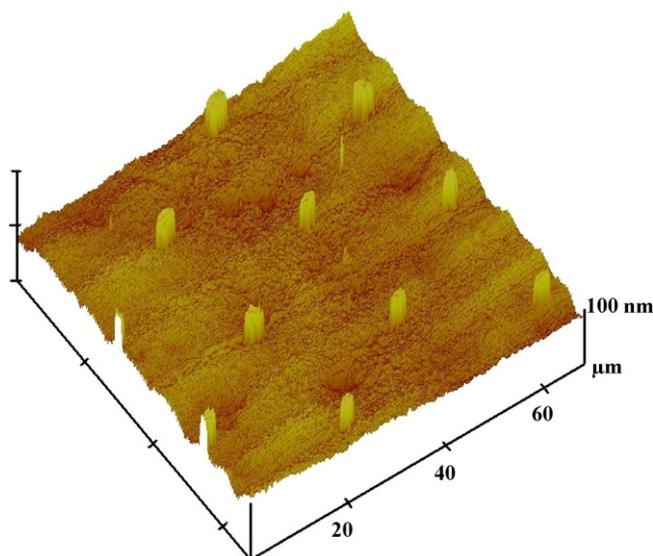


Fig. 6. Three-dimensional AFM image of patterns in the phase change film after NaOH etching.

The results above show that both laser fluence and laser wavelength affect the feature size. The laser fluence should be small enough to overcome the focus size limitation of the microlens in order to achieve nanosized features. However, the feature size is still limited by the laser wavelength and cannot be made smaller by using an Nd:YAG laser. To achieve higher resolution, a laser with a shorter wavelength or a shorter pulse duration laser should be used. This will be investigated in future experiments.

5. Conclusions

A large number of crystalline features are fabricated in a phase change film simply and uniformly by irradiation with an Nd:YAG laser through a microlens array, with the fundamental and second harmonic components of the Nd:YAG laser acting as the light source. The feature size produced by shorter wavelength laser irradiation is smaller, and the size increases with laser fluence linearly for both laser wavelengths. At low laser fluence, the feature size formed is much smaller than the calculated minimum focus beam size and the smallest feature size achievable in this work is around 355 nm, produced by laser irradiation at a wavelength of 532 nm. By making use of shorter laser wavelength or shorter laser pulse, it is expected that much smaller pattern size in the phase change film can be created. This would therefore be a useful technique for high capacity optical data storage.

Acknowledgement

The authors would like Ms. Doris Ng for her useful advice and the financial support of Agency for Science, Technology and Research (A*Star) in Singapore.

References

- [1] Y. Kageyama, H. Iwasaki, M. Harigaya, Y. Ide, *Jpn. J. Appl. Phys.* 35 (1996) 500.
- [2] N. Miyagawa, Y. Gotoh, E. Ohno, K. Nishiuchi, N. Akahira, *Jpn. J. Appl. Phys.* 32 (1993) 5324.
- [3] T. Shintani, K.N.S. Hosaka, A. Hirotsune, M. Terao, R. Imura, K. Fujita, M. Yoshida, S. Kammer, *Ultramicroscopy* 61 (1995) 285.
- [4] M. Petran, M.D. Egger, *Science* 157 (1967) 305.
- [5] A. Ichihara, T.T.K. Isozaki, Y. Sugiyama, Y. Kosugi, K. Mikuriya, M. Abe, I. Umeda, *Bioimages* 42 (1996) 57.
- [6] J. Bewersdorf, R. Pick, S.W. Hell, *Opt. Lett.* 23 (1998) 655.
- [7] K. Fujita, O. Nakamura, T. Kaneko, M. Oyamada, T. Takamatsu, S. Kawata, *Opt. Commun.* 174 (2000) 7.
- [8] M.-H. Wu, K.E. Paul, G.M. Whitesides, *Appl. Opt.* 41 (2002) 2575.
- [9] J. Kato, N. Takeyasu, Y. Adachi, H.B. Sun, S. Kawata, *Appl. Phys. Lett.* 86 (2005) 044102.
- [10] T. Nonaka, G.O.Y. Toriumi, Y. Mori, H. Hashimoto, *Thin Solid Films* 370 (2000) 258.
- [11] N. Yamada, T. Matsunaga, *J. Appl. Phys.* 88 (2000) 7020.
- [12] B. Besold, N. Lindle, *Pure Appl. Opt.* 6 (1997) 691.
- [13] H.F. Talbot, *Phil. Mag.* 9 (1836) 401.
- [14] N. Yamada, E.O.K. Nishiuchi, N. Akahira, *J. Appl. Phys.* 69 (1991) 2849.