

Theoretical analysis for temperature dependence of laser-induced damage threshold of optical thin films

K Mikami^{1,2}, S Motokoshi³, T Somekawa³, T Jitsuno¹, M Fujita³, KA Tanaka²,
and H Azechi¹

¹ Institute of Laser Engineering, Osaka University, 2-6 Yamada-oka, Suita, Osaka 565-0871 Japan

² Graduate School of Engineering, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871 Japan

³ Institute for Laser Technology, 1-8-4 Utsubo-honmachi, Nishi-ku, Osaka 550-0004 Japan

E-mail: kmikami@ile.osaka-u.ac.jp

Abstract. The temperature dependence of the laser-induced damage threshold on optical coatings was studied in detail for laser pulses from 123 K to 473 K at different temperatures. The laser-induced damage threshold increased with decreasing temperatures when we tested long pulses (200 ps and 4 ns). The temperature dependence, however, was reversed for pulses shorter than a few picoseconds (100 fs testing). We propose a scaling model with a flowchart that includes three separate processes: free-electron generation, electron multiplication, and electron heating. Furthermore, we calculated the temperature dependence of laser-induced damage thresholds at different temperatures. Our calculation results agreed well with the experimental results.

1. Introduction

Next generation high-power laser systems are expected to develop as one of the essential tools in several scientific and industrial fields. The Yb:YAG laser medium for cryogenically cooled laser systems is a typical material [1,2] and has been studied in ILE Osaka University [3-5]. In particular, the laser system using Yb:YAG ceramic is planned as a laser fusion driver.

The output power of a high-power laser system is limited by the laser-induced damage threshold (LIDT) because damaged optics cannot provide the required performance. Improvement of the LIDT is important to enhance the output power of a high-power laser system. A laser-induced damage mechanism (LIDM) has been studied for over 40 years [6]. A typical study of pulse width dependence of the LIDT was reported by Stuart *et al.* in 1995 [7]. That study revealed a new scaling law, $\tau^{1/2}$ scaling and the role of pulse width in the LIDM. In particular, a balance between linear and nonlinear phenomena was mentioned as an essential parameter to provide the scaling, which is an important consideration for high-intensity laser system design.

The role of temperature in the LIDM is extremely important for cryogenically cooled laser systems, and several studies for temperature dependence of LIDT have been reported [8-13]. However, the theoretical approach for the effect in the LIDM is insufficient to discuss to estimate the LIDT measured by different pulse widths at different temperatures.

¹ To whom any correspondence should be addressed. E-mail: kmikami@ile.osaka-u.ac.jp



In this study, we demonstrated the measurement of the LIDT at different temperatures with different pulse widths; furthermore, we suggested a model to explain the data.

2. Experiment

2.1. Experimental setup

We evaluated the temperature dependence of the LIDT using a single-mode Nd:YAG laser (wavelength 1064 nm, pulse width 4 ns, single shot) and a Ti:Sapphire laser (wavelength 800 nm, pulse width 100 fs, 2 ps, and 200 ps, repetition frequency 10 Hz). Figure 1 shows the experimental setup. The irradiation energy of the laser pulse was adjusted with a half-wave plate and a polarizer. The mechanical shutter was used to pick up a pulse from the Ti:Sapphire laser. The laser pulse was focused by a lens ($f = 200$ mm), and the spot sizes were defined by the $1/e^2$ peak intensity and were $80\text{ }\mu\text{m}$ ($\lambda = 800$ nm) and $160\text{ }\mu\text{m}$ ($\lambda = 1064$ nm) in diameter. The spot size was measured by image transferring on a sample surface to a CCD camera using a lens ($f = 600$) with $3\times$ magnification. The sample was held by a Cu holder and set in a vacuum chamber (~ 5 Pa) to prevent water condensation at low temperature. The incident angle was 0° . The sample's temperature was adjusted with liquid N_2 and a heater, and monitored with a Pt resistance temperature detector attached to the sample holder.

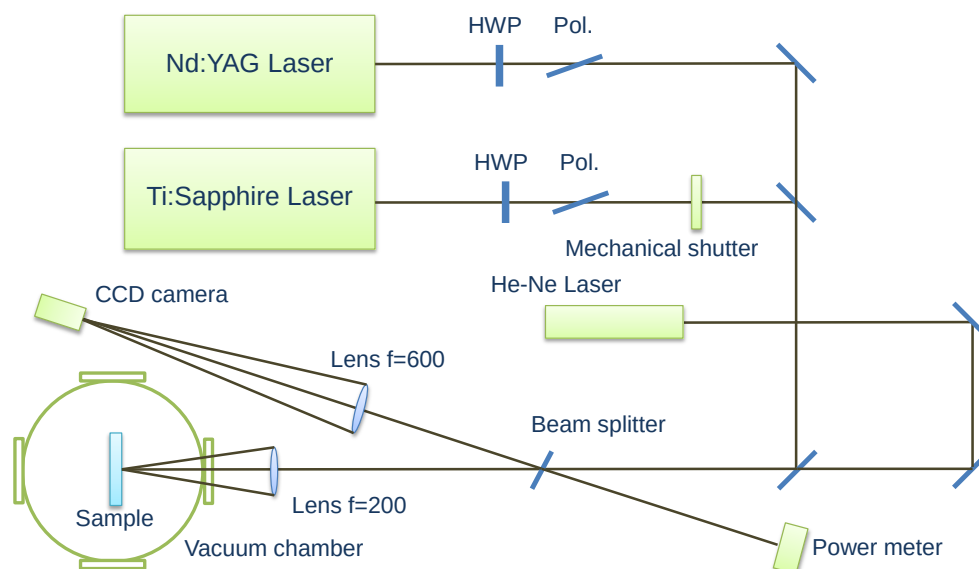


Figure 1. Experimental layout

2.2. Determination method for LIDT

In this study, we employed the N-on-1 test method. A fixed site was irradiated by laser pulses. The fluence was gradually increased until damage occurred. The total shot number of laser pulses was approximately 15 shots at each test. The laser-induced damage was detected by plasma emission and transfer of the surface image with a co-aligned He-Ne laser. In the method for damage detection, we could detect a damage site with a minimum diameter of $2\text{ }\mu\text{m}$. The measured LIDT was tested with three average values at the same experimental conditions.

2.3. Experimental sample

Six types of single-layer coatings (SiO_2 , Al_2O_3 , HfO_2 , ZrO_2 , Ta_2O_5 , and MgF_2) were prepared on optical polished silica glass substrates, with a root mean square roughness $< 0.8\text{ nm}$, by electron beam evaporation. The optical thickness of the single layers had a quarter wavelength ($nd = \lambda/4$) at 800 nm.

2.4. Experimental result

Figure 2 shows the temperature dependence of the LIDT for (a) SiO₂, (b) Al₂O₃, (c) HfO₂, (d) ZrO₂, (e) Ta₂O₅, and (f) MgF₂ single-layer coatings. The vertical axis indicates the normalized LIDT with the LIDT at room temperature. Table 1 shows the LIDT at room temperature. The LIDT for all samples decreased with decreasing temperature in 100 fs testing. With the 2 ps laser pulse, however, the dependence of some materials showed an increase in the LIDT with decreasing temperature. Additionally, under 200 ps and 4 ns laser pulses, the LIDT exhibited the opposite trend. The slope of the LIDT for each coating material was changed for the pulse width around a few picoseconds.

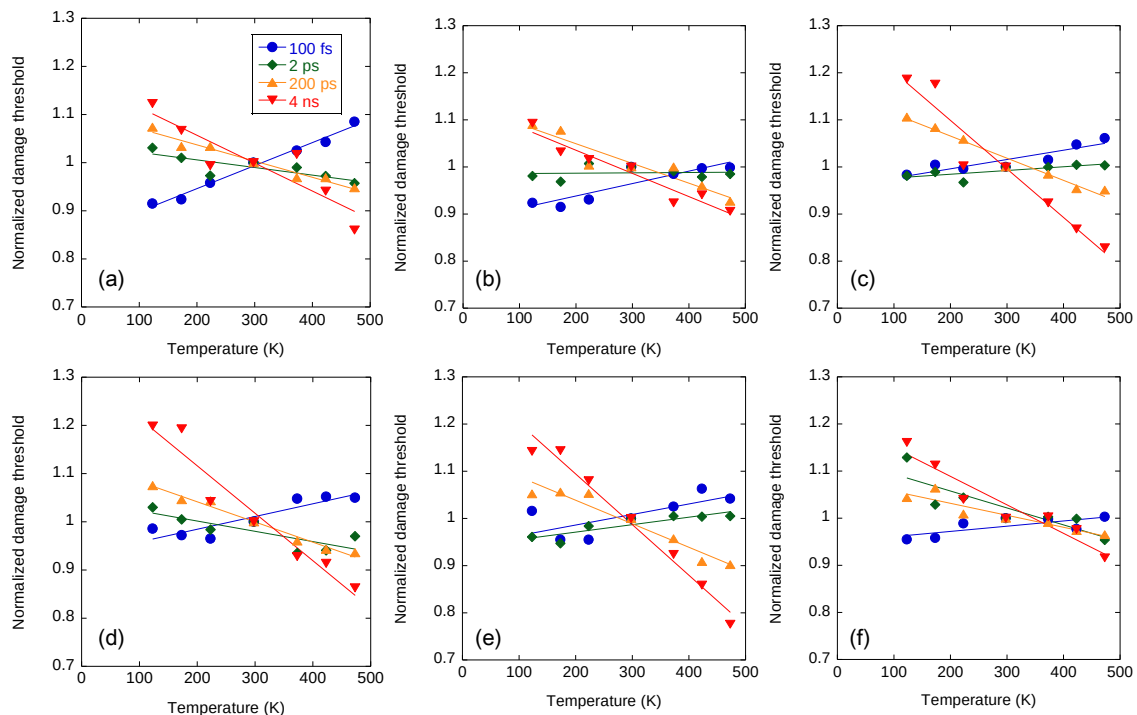


Figure 2. Temperature dependence of LIDT for (a) SiO₂, (b) Al₂O₃, (c) HfO₂, (d) ZrO₂, (e) Ta₂O₅, and (f) MgF₂ single-layer coatings

Table 1. LIDT at room temperature

Pulse width	LIDT at room temperature (J/cm ²)					
	SiO ₂	Al ₂ O ₃	HfO ₂	ZrO ₂	Ta ₂ O ₅	MgF ₂
100 fs	0.55	0.39	0.36	0.35	0.30	0.65
2 ps	2.0	1.43	1.4	1.44	1.4	1.5
200 ps	5.6	437	4.5	4.5	4.3	5.3
100 fs	22	22	9.1	10	7.1	22

3. Quantitative analysis

3.1. Theoretical model

A flowchart was fabricated using the LIDM to calculate the LIDT, as shown in Fig. 3. The damage formation was constructed with three processes: free-electron generation, electron multiplication, and electron heating. The route in the flowchart was branched because of the irradiated pulse width. In the short-pulse region, with no more than a few picoseconds, the high intensity causes multiphoton ionization as a nonlinear optical phenomenon. By contrast, the lower intensity of the short pulse leads to linear optical phenomena, *i.e.*, photoionization and phonon–electron interaction, in the long-pulse region, which lasts more than a few picoseconds.

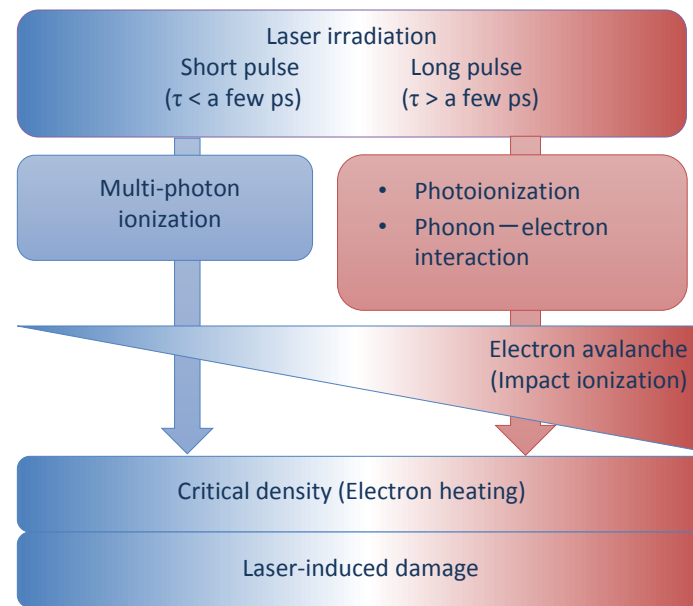


Figure 3. Flow chart using LIDM for calculation LIDT

Multiphoton ionization [7] is the transfer of electrons from the valence band to the conduction band with two or more photon absorptions (*multiphoton* absorption). Photoionization [14] is the transfer of electrons from the defect and/or impurity state to the conduction band with single photon absorption. Phonon–electron interactions, an important process at long pulse widths, were studied using the energy gain rate G and average energy loss rate L of an electron and the average electron model [11]. The number densities of generated electrons by multiphoton ionization, n_m , and phonon–electron interaction, n_i , can be calculated using Eqs. (1) and (2) [7,11].

$$n_i = \frac{G - L}{E_g V} = \frac{(e^2/m)\tau_{s(\varepsilon)}E^2 - \hbar\omega}{E_g V} \quad (1),$$

$$n_m = \sigma_4 N_s \left(\frac{c\varepsilon E^2}{2\hbar\nu} \right)^4 \left(\frac{\pi}{\ln 2} \right)^{0.5} \frac{\tau}{4} \quad (2),$$

where E_g is the band gap energy, V is the volume, e is the charge of electron, m is the electron mass, $\tau_{s(\varepsilon)} = 30$ fs is the collisional time [7], E is the electric field, ω is the optical phonon frequency [15], $\sigma_4 = 2 \times 10^{-144} \text{ cm}^8\text{s}^3$ is the cross section of four-photon absorption [7], N_s is the atomic density, c is the light speed, ε is the dielectric constant, and ν is the laser frequency. In this study, we assumed V to be a product of the irradiated spot size and sample thickness. The Rayleigh length is better than the assumption in the calculation when the sample thickness is longer than the Rayleigh length. Additionally, the number density of a free electron from photoionization, n_p , was estimated to be 10^{-14} cm^{-3} , assuming an impurity concentration on the order of 0.01 ppm to 1 ppm.

The generated free electrons are multiplied by impact ionization and electron avalanche. The process has a strong tendency to be dominant under irradiation with a long pulse laser. The multiplied number density is calculated using the number density of the generated free electron, volume V , pulse width τ , and multiple rate β . An equation to calculate the multiple rates has been reported:

$$\beta = A \frac{E^2}{E_g \rho} \quad (3),$$

where $A = 17$ is an arbitrary constant and ρ is the electrical resistivity [16]. Eq. (3) can be used under the assumption that the multiplication of the free electrons is exponential because of adequate

acceleration time (pulse width). When the acceleration time is not adequate, that is, no more than a few picoseconds, Eq. (3) is replaced by Eq. (4) [17],

$$\beta' \tau V = 2^{\tau/\tau_{s(\varepsilon)}} \quad (4).$$

Finally, the multiplied free electrons are heated with a laser light. The number density under electron heating is discussed with Bose–Einstein statistical mechanics. The number density is defined as critical density n_{cr} and described as Eqs. (5) and (6) [18],

$$n_{cr} = \frac{p}{4\pi e^{*4}} \frac{\hbar \omega}{\tau_{s(\varepsilon)} (2N_w + 1)} \quad (5),$$

$$N_w = \left\{ \exp \left(\frac{\hbar \omega}{k_0 T} \right) \right\}^{-1} \quad (6),$$

where $p = (2m\varepsilon_e)^{1/2}$ is the momentum of free electrons, $e^* \sim e(1/\varepsilon)^{1/2}$ is the effective electron charge, k_0 is the Boltzmann constant, and T is the temperature.

The conditional expressions were described as Eqs. (7) and (8), and calculated using Eqs. (1)–(6)

$$n_{cr} = \beta \tau V (n_p + n_i) \quad (7),$$

$$n_{cr} = \beta' \tau V n_m \quad (8).$$

Eqs. (7) and (8) can give the electric field that triggers the laser-induced damage. The given electric field is converted to intensity and fluence, that is, to the LIDT.

3.2. Calculation results

The previous discussion enables us to model the temperature dependence of the LIDT. Figure 4 shows the measured and calculated temperature dependence of the LIDT for SiO₂ coatings at 100 fs and 4 ns. The solid line in Fig. 4 shows the results of the calculation with Eq. (7). The dashed line in Fig. 4 shows the results of the calculation with Eq. (8). The calculated results are in good agreement with the experimental results.

The model can further reveal the key parameter for the temperature dependence of the LIDT. Table 2 shows the calculated results using the proposed model for the variation when the temperature is changed from 473 K to 123 K.

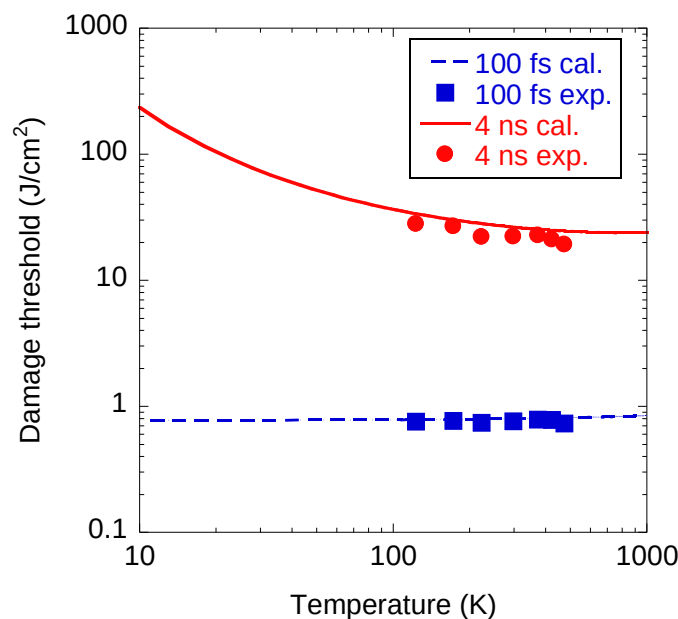


Figure 4. Measured and calculated temperature dependence of LIDT for SiO₂ coating.

When the LIDT in a short-pulse region is calculated by Eq. (8), the LIDT decreases with decreasing temperature. This is because the critical density at 123 K is 1.2 times lower than that at 473 K. That is, the free electrons at a higher temperature have a lower density than those at a lower temperature, and therefore, the LIDT decreases. When the LIDT in a long-pulse region is calculated by Eq. (7), however, the LIDT increases with decreasing temperature. The number density of free electrons by phonon–electron interaction at 473 K increases by 1.2 times because of the inhibited optical phonon frequency. However, the variation in phonon–electron interaction is smaller than the variation in multiple rates. Figure 5(a) shows the temperature dependence of electrical resistivity for SiO₂ [17], and Fig. 5(b) shows the calculated multiple rate. The temperature dependence of the LIDT in long-pulse testing is decided by that of the multiple rate, *i.e.*, the temperature dependence of the electrical resistivity. The electron avalanche in the long-pulse region can become more dominant in the LIDM than in the short-pulse region. The variation in the trend of the temperature dependence of the LIDT will be decided because of the electron avalanche process.

Additionally, the model for the temperature dependence of the LIDT may suggest a method of producing extremely high LIDT optics, at least several times higher than conventional optics, because the calculated LIDT (solid line in Fig. 4) exponentially increases with decreasing temperature. The LIDT at a low temperature can be improved mainly by controlling the electrical resistivity. If the temperature dependence of each parameter, particularly electrical resistivity, is fixed at a cryogenic condition, we might obtain extremely high LIDT optics.

Table 2. Calculation result for variation (temperature from 473 K to 123 K)

Process	Variation
Free-electron generation	Number density of free electron by photoionization
	No change
	Number density of free electron by phonon-electron interaction
	1.2 times increase
Electron multiplication	Number density of free electron by multi photon ionization
	No change
	Impact ionization
	No change
Electron multiplication	Multiple rate in electron avalanche
	2 times decreased
Electron heating	Critical density
	1.2 times decrease

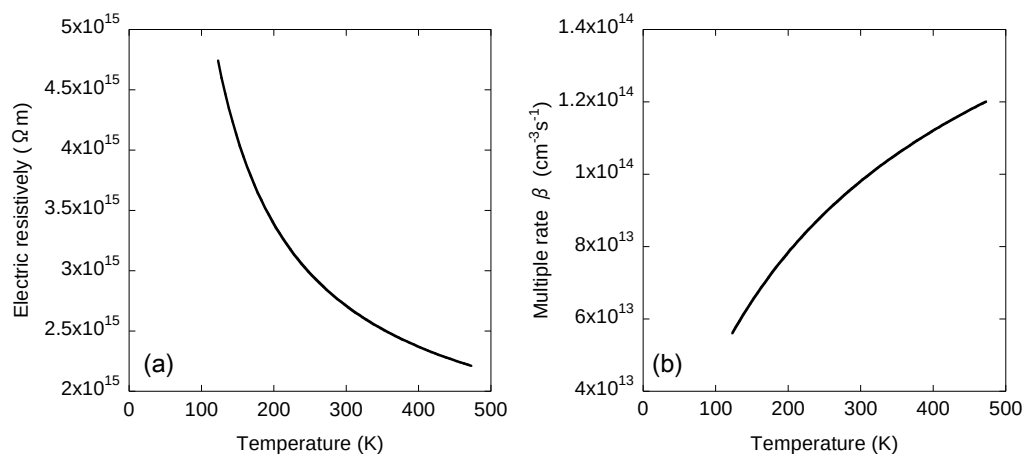


Figure 5. (a) Temperature dependence of the electric resistivity of SiO₂ and (b) temperature dependence of electron multiple rate.

4. Conclusion

In conclusion, we measured the temperature dependence of the LIDT of optical coatings at different pulse widths. The trend of the LIDT temperature dependence varied because of the irradiated pulse width. The calculated results obtained using the proposed model agreed well with the measured results. The electrical resistivity, which is related to the electron avalanche, is an essential parameter affecting the temperature dependence. To improve the LIDT of optical materials, it is particularly important to evaluate the temperature dependence of the electrical resistivity.

We may also propose a method of producing extremely high LIDT optics, at least several times higher than conventional optics, because the calculated LIDT (solid line in Fig. 4) exponentially increases with decreasing temperature. LIDT at a low temperature can be improved mainly by controlling the electrical resistivity. We can realize this new type of optics by combining existing optics and a cryostat, where the mirror substrate can be cooled down to either 77 K or 2 K with liquid N₂ or He. The new optics will have the potential to demonstrate high power that has not been achieved to date.

5. References

- [1] Broun B C, Bruno T M and Singley M 2010 *Opt. Express* **18** 16573
- [2] Broun B C, Singley J M, Yager E, Kowalewski, Guelzow J and Kuper J W 2008 *Proc. SPIE* **6952** 69520K
- [3] Kawanaka J, Tokita S, Nishioka H, Fujita M, Yamakawa K, Ueda K and Y Izawa 2005 *Phys. Laser* **15** 1306
- [4] Furuse H, Kawanaka J, Takeshita K, Miyana N, Saiki T, Imasaki K, Fujita M and S Ishii 2009 *Opt. Lett.* **34** 3439
- [5] Kawanaka J, Takeushi Y, Yoshida A, Parce S J, Yasuhara R, Kawashima T and Kan H 2010 *Phys. Laser* **20** 1079
- [6] Manekov A A 2008 *Proc. SPIE* **7132** 713202
- [7] Stuart B C, Feit M D, Rubenchik A M, Shore B W Perry M D 1995 *Phys. Rev. Lett.* **74** 2248
- [8] Mikami K, Motokoshi S, Fujita M, Jitsuno T, Kawanaka J and Yasuhara R 2009 *Proc. SPIE* **7504** 75041R
- [9] Mikami K, Motokoshi S, Fujita M, Jitsuno T, Kawanaka J and Yasuhara R 2010 *J. Phys.: Conf. Ser.* **244** 032023
- [10] Mikami K, Motokoshi S, Fujita M, Jitsuno T and Tanaka K A 2012 *Proc. SPIE* **8190** 81900A
- [11] Manenkov A A 1978 *Natl. Bur. Stand. (U.S.) Spec. Publ.* **541** 455
- [12] Markle L D and Kitriouns D 1988 *Phys. Rev. B* **38** 1473
- [13] Plantanak J R, Walser R M and Becker M F 1987 *Natl. Bur. Stand. (U.S.) Spec. Publ.* **752** 216
- [14] Braunlich P and Schmid A 1975 *Appl. Phys. Lett.* **26** 150
- [15] Worlock J M and Fleury P A 1967 *Phys. Rev. Lett.* **19** 1176
- [16] Roy S and Chakravorty 1995 *J. Mater. Res.* **9** 2314
- [17] Seitz F 1949 *Phys. Rev.* **76** 1376
- [18] Frohlich H and Paranjape B V 1956 *Proc. Phys. Soc. B* **69** 21

Acknowledgments

This study was funded by a Grant-in-Aid for Research Fellow from the Japan Society for the Promotion of Science (to K Mikami, Grant No. 25-275).