Thermal quench of ion-beam induced luminescence of Er₂O₃

高温加熱によるEr₂O₃のイオンビーム誘起蛍光の消光現象

<u>D. Kato</u>, H.A. Sakaue, I. Murakami, T. Tanaka, T. Muroga and A. Sagara 加藤太治, 坂上裕之, 村上泉, 田中照也, 室賀健夫, 相良明男

National Institute for Fusion Science 322-6, Oroshi-cho, Toki, Gifu 509-5292, Japan 核融合科学研究所 〒509-5292 岐阜県土岐市下石町322-6

 Er_2O_3 is a prospective material used for electric insulation and tritium permeation barrier of advanced blanket systems with liquid breeders and coolants, e.g., Li, Fliebe, and Li-Pb. Ion-beam induced luminescence of Er^{3+} in the oxide crystal bombarded by energetic Ar^+ ion-beam was measured at elevated temperatures up to 873 K. Quenching of the luminescence band in 640-690 nm was found at high temperatures. Population distribution in the upper Stark levels of the luminescence agrees almost with Boltzmann distribution at target temperatures. A characteristic lifetime of the upper levels measured from the emission line broadening decreases as the temperature increases.

1. Introduction

This study is undertaken to develop optical methods for *in-situ* characterization of radiation-induced defects in Er_2O_3 by energetic ion bombardment. The characterization is important for qualification of Er_2O_3 coatings as electric insulation of Li/V-alloy blanket systems [1] and as hydrogen permeation barriers [2].

A potentially useful luminescence band in 640 – 690 nm is identified as $4f^{11} {}^{4}F_{9/2} \rightarrow {}^{4}I_{15/2}$ transition of Er^{3+} at C_2 cation sites. It has been demonstrated that ion bombardment on a plasma splay coating sample [3] and a sintered bulk sample [4] quenches the luminescence band. This is explained by a model assuming ion induced decrystallization of the target [4, 5].

The blanket material will be used at elevated temperatures in fusion reactors. In the present work, we address the defect characterization at the elevated temperatures by the ion-beam induced luminescence measurements with a heated Er_2O_3 sample.

2. Experimental setup

The Er_2O_3 sample (high purity (3N) sintered plate, 15mm × 15mm wide and 1mm thick, TEP CORP.) is mechanically attached on a micro ceramic heater equipped with a thermocouple (25mm × 25mm wide, MS-1000R, SAKAGUCHI E.H VOC CORP.). Ceramic fiber papers are used to thermally insulate the ceramic heater from a target holder on which the ceramic heater is set. Temperature control of the heater is performed automatically with the aid of a thermoregulator (SCR-SHQ-A, SAKAGUCHI E.H VOC CORP.). The temperature of the target sample is deduced assuming thermal equilibrium with the ceramic heater.

Ion-beam induced luminescence of the target sample is measured with an experimental apparatus at NIFS consisting of an ion-beam source, a collision chamber, and a UV-visible spectrometer equipped with a Peltier cooled CCD camera. The ion source is a part of medium current ion implanter (ULVAC IM-200MH-FB) originally used for semiconductor production (Freeman-type). Ion-beams extracted from the Freeman ion source are introduced into the collision chamber after analyzing the mass to charge ratio by a magnet. In the present experiments, we could use Ar⁺ ion beam of about 10 µA in current at 35 keV in kinetic energy.



Fig. 1 Collision chamber and visible spectrometer [4].

Photon emission is measured at a right angle to the ion-beam axis. The sample surface is leaned 45 degrees toward the line of sight so that the photons emitted in an area of the ion-beam spot are collected by a condenser lens focused on a slit of the spectrometer as shown in Fig. 1. Wavelength calibration was done by using emission lines of H and He discharge tubes.

3. Results and discussion

Figure 2 shows luminescence spectra induced by Ar^+ ion irradiation at five temperatures from 373 K to 773 K. The luminescence intensities in 640 - 690 nm decrease as the target temperature increases. In contrast, emission lines at shorter wavelengths than 640 nm, which are assigned to the lines of sputtered Er neutrals, become stronger as the temperature increases.



Fig. 2 Ion-beam induced luminescence of Er₂O₃.

Strong lines A, B, and C are assigned to the lines due to transitions between the Stark levels: ${}^{4}F_{9/2} M = 1/2$, 9/2, $9/2 \rightarrow {}^{4}I_{15/2} M = 9/2$, respectively. Intensity ratios among the lines A, B and C were analyzed to investigate population distributions in the upper Stark levels. Each line intensity was evaluated by fitting the measured spectrum to triple Lorentz functions. The ratios can be fitted to Arrhenius equations. Exponents of the Arrhenius equations are consistent with energy differences among the Stalk levels. This indicates that the population distribution in the upper Stalk levels is in the thermal equilibrium at the ambient temperature.

Figure 3 shows a characteristic lifetime of the upper levels which are evaluated from the line broadening. The lifetime decreases as the temperature increases as shown in the figure. This may indicate that non-radiative depopulation of the upper levels is facilitated at high temperatures.



Fig. 3 Lifetime of ion-beam induced luminescence due to ${}^{4}F_{9/2} \rightarrow {}^{4}I_{15/2}$ transition of Er^{3+} in Er_2O_3 .

It is known that photo luminescence at 1.54 μ m of Er³⁺ doped in Si quenches at the room temperature [6]. However, co-doping of other impurities shifts the quenching temperature significantly higher. In this case, electron-hole recombination at the impurity levels in the Si band gap play a primary role to excite the luminescence [7]. The quenching at high temperatures observed in the present work may also be associated with some defect levels introduced by energetic ion bombardment.

Acknowledgments

DK is grateful to supports by Fusion Engineering Research Project in NIFS (UFFF029) and NIFS General Collaboration Projects (NIFS12KBAF006, NIFS14KLEF014).

References

- T. Muroga et al.: J. Nucl. Mater. 367–370 (2007) 780.
- [2] D. Levchuk et al.: J. Nucl. Mater. 367–370 (2007) 1033.
- [3] T. Tanaka et al.: J. Nucl. Mater. 417 (2011) 794.
- [4] D. Kato et al.: Plasma Fusion Res. 7 (2012) 2405043.
- [5] D. Kato et al.: "Optical properties of Er_2O_3 damaged by ion-beam bombardment", The 9th Joint Conference for Fusion Energy (Kobe Convention Center, 28-29 June, 2012) 29A-62p.
- [6] A. Polman, J. Appl. Phys. 82 (1997) 1.
- [7] F. Priolo et al., J. Appl. Phys. 78 (1995) 3874.