Effect of ultrasonic oscillatory stress on deformation luminescence of X-irradiated KCl : Eu2 +

Effect of ultrasonic oscillatory stress on deformation luminescence of X-irradiated $KCl:Eu^{2+}$

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Abstract

Luminescence has been observed during deformation of X-irradiated alkali halide crystals. And also, it has been observed at fracture or under superimposition of ultrasonic oscillation. These phenomena are useful for studying the deformation of crystals. We have investigated luminescence during the deformation of X-irradiated $KCl:Eu^{2+}$ and under superimposition of oscillation.

The luminescence intensity rapidly increases at the beginning of plastic deformation. The intensity roughly remains constant for a while and is then followed by gradual decreasing. The variation of luminescence intensity seems to correspond to the deformation stage of the crystal. The spectrum of deformation luminescence (DL) has a peak of 420nm due to Eu^{2+}. Superimposition of oscillation during plastic deformation of crystals causes a decrease in stress along with additional luminescence (sonoluminescence SL). The SL intensity also appears to correspond to the deformation stage. SL/DL is found to be proportional to stress decrement due to superimposition of ultrasonic oscillation irrespectively of strain. DL has been found to occur mainly at the intersection of slip bands.

Keywords: X-irradiation; Deformation luminescence; $KCl:Eu^{2+}$; oscillation

1. Introduction

There are many reports on mechanoluminescence (ML) for irradiated alkali halide crystals (Butler, 1966; Zakrevskii *et al*., 1995). It has been reported that the ML

intensity is proportional to the F center concentration of the crystal after X- or γ -irradiation, while its spectra corresponds to that of thermoluminescence. It is then believed that the ML is attributed to the recombination of the electrons released from F^- centers with hole centers (Hayashiuchi *et al*.,1990).

Recently, at fracture under three point bending, fractoluminescence (FL) of X-irradiated KCl:Ca^{2+} single crystals are proportional to the third power of the fracture load. It is suggested that FL must depend on the number of intersections of moving dislocation with forest dislocation (Ohgaku *et al*. 2002).

On the other hand, sonoluminescence (SL) has been observed in X-irradiated KCl crystals. It has been reported that there is critical strain amplitude above which the luminescence becomes notable and the internal friction indicates an abrupt increase (Miyake and Futama, 1982). The unpinning process of dislocation has been considered to cause the phenomenon and the process annihilates defects that have pinned the dislocation.

The aim of this work is to investigate the influence of deformation process and oscillation on luminescence of irradiated KCl:Eu^{2+} single crystals during plastic deformation.

2. Experimental procedure

Samples were cleaved out of an ingot of KCl single crystals doped with Eu^{2+} which were grown from the melt of reagent powder by Bridgeman method, followed by annealing at 1030 K for 24 h and cooling at 40 K/h to room temperature. The size of samples is 2x2x5 mm^3. The samples were exposed to X-ray from a W-target being operated at 40 kV and 30 mA for 1 min.

Fig.1 shows the schematic for the apparatus. The sample was plastically deformed by compression in the <100> direction along the largest axis at the cross-head speed of 20 μm min^{-1} using an Instron 4465 testing machine. Ultrasonic oscillatory stress with the frequency of 20 kHz was applied to the sample during plastic deformation in the same direction as compression. Superimposition of oscillation was performed intermittently at various stress amplitudes, which was monitored by a piezoelectric transducer set between a specimen and a supporting rod. The oscillatory stress of the specimen was homogeneous because the specimen size was much smaller than the

wavelength.

Luminescence was observed using a photomultiplier (Hamamatsu R928). Luminescence intensity and load signals were recorded on a computer and then the data was processed. Luminescence patterns on a sample were recorded using CCD camera (Hamamatsu PMA-100). All experiments were carried out at room temperature.

3. Results and discussion

Fig.2 shows absorption spectra of X-irradiated KCl:Eu^{2+} for various exposure time. The spectra have three peaks. The peek of 580nm is due to F-center and the others (243 and 343nm) are due to Eu^{2+}. The height of the peak due to the F-center increases with increasing exposure time but that due to Eu^{2+} seems to decrease taking account of base absorption line. The F-center concentration is 8.3ppm using Smakura's formula and that of is 67ppm according to Hernandez *et al*. for 1min exposure.

Luminescence is observed in X-irradiated KCl:Eu^{2+} during plastic deformation (deformation luminescence DL) in Fig.3, although no luminescence was observed in a non-irradiated sample. This suggests that the color center induced by X-irradiation is responsible for luminescence during deformation as well as thermoluminescence.

It is shown in Fig.3 that static stress (a) decreases and excess luminescence intensity (b) is added to the DL intensity, which forms the base line of luminescence without oscillation, when the oscillatory stress is superimposed. This additional luminescence is here named sonoluminescence (SL). The DL intensity in Fig.3 (b) rapidly increases at the beginning of deformation and roughly remains constant for a while, followed by its gradual decrease. The variation of DL intensity seems to correspond to the deformation stage of the crystal.

The SL intensity increases sharply at the beginning of oscillation and reaches a maximum that is given by the stress amplitude. Then the SL intensity gradually decreases to a constant value. When oscillation is turned on, dislocation should abruptly move a large distance because the stress exerted on dislocation increases for a moment although it is not observed as static stress. This leads to the initial sharp increase of SL intensity. The SL intensity is considered to reach a constant when the average velocity of dislocation under oscillation is equal to that without oscillation. The SL intensity is hereafter given by its maximum value under superimposition of oscillation. The stress

decrement ($\Delta\tau$) as well as SL intensity increases with increasing stress amplitude. This means that oscillation induces stronger luminescence.

Since the luminescence due to dislocation motion is considered to be proportional to the area where dislocation sweeps, oscillating dislocation must sweep a larger area per unit time. The area swept by the oscillating dislocation for unit time may be proportional to the stress amplitude if the dislocation bowing between pins is small and internal friction does not depend on the stress amplitude. Then, the SL intensity is expected to be proportional to the stress decrement ($\Delta\tau$) because $\Delta\tau$ is also approximately proportional to the stress amplitude (Ohgaku and Takeuchi, 1987).

In order to investigate the relation between $\Delta\tau$ and SL in the given internal structure of a sample, the relation of $\Delta\tau$ and SL/DL with strain is obtained in Fig.4 at the several magnitude of stress amplitude. The values of SL/DL and $\Delta\tau$, which are read at the strain of 8, 12, 16, 20, and 24% from Fig.4, are plotted in Fig.5. The SL/DL is seen from the figure to be proportional to $\Delta\tau$ irrespective of strain although both SL and DL decrease with increasing strain as shown in Fig.3. This means that both DL and SL depend on the concentration of remaining F-center by the same manner and the stress amplitude in this test is not enough to cause amplitude dependent internal friction (Nakamura *et al*. 2006).

It is an interesting problem what mechanism cause the deformation luminescence of X-irradiated KCl:Eu^{2+}. Fig.6 shows the collection of luminescence spectra under superimposition of oscillation at each deformation stage. Each spectrum has a peak of 420nm irrespectively of strain. The peak of 420nm is the same peak as that obtained by optically stimulated luminescence measurements of KCl:Eu^{2+} (Nanto *et al*., 1998). So, DL and SL is explained as follows.

It should be noticed that the absorption band at 580nm of F-centers increases and that at 243 and 343 of Eu^{2+} decreases when exposure time increases, as shown in Fig.2. Then, it is likely that part of the free electrons and holes created by X-irradiation are trapped at anion vacancies to produce the F-centers and at Eu^{2+} to produce Eu^{3+} ions, respectively. During deformation or dislocation motion, electrons released from the F-centers combine with the Eu^{3+} ions leading to the exited Eu^{2+} ions from which the 420nm DL emitted. This process obey the following reaction,

$$Eu^{3+}+e^{-} \rightarrow (Eu^{2+})^{*} \rightarrow Eu^{2+}+h\nu(420\ \mathrm{nm}),$$

where e^{-} indicates free electrons and $(Eu^{2+})^{*}$ is the divalent europium exited states.

Consequently, the transition from the exited states of divalent europium to the ground states results in the 420 nm luminescence.

It has not been clear how electrons are carried from F-centers to Eu^{3+} ions. Whether electrons are exited by the thermal spike of dislocation depinning from obstacles or they are directly carried by mobile dislocation. Anyway, dislocation motion causes the transference of electrons from F-centers to Eu^{3+} ions and causes luminescence at Eu^{2+} ions.

Fig.7 shows the photographs of luminescence pattern on the surface of a sample at each deformation stage together with the variations of stress and DL. The DL intensity on the photograph as well as on the graph of DL vs. strain appears to increase at the beginning of deformation and decrease gradually in the second or third deformation stage. Flash happened when a sample seemed to crack during deformation. And there appears the intense flash at the fracture as shown in the figure. The luminescence pattern changes as plastic deformation proceeds. The luminescence appears to take place along the slip bands and the most intense luminescence occurs in the intersection area of slip bands. This leads to the consideration that dislocation cutting causes deformation luminescence. The dislocation cutting makes jog or kink on the dislocation line. The jog or kink may be related to luminescence during deformation of crystals. Since the jogs on edge dislocation line have electric charges the edge dislocation may carry electric charges.

4. Conclusion

Luminescence is observed neither during deformation nor under superimposition of oscillation for non-irradiated $KCl:Ca^{2+}$ crystals. However, sonoluminescence is observed under superimposition of oscillation during plastic deformation of X-irradiated $KCl:Ca^{2+}$ as well as without oscillation.

The DL intensity rapidly increases at the beginning of deformation and keeps roughly constant for a while, followed by its gradual decrease, depending on the deformation stage.

The SL intensity increases sharply at the beginning of oscillation and reaches a maximum that is given by the stress amplitude. And it gradually decreases to a constant value. The SL/DL is found to be proportional to $\Delta\tau$ irrespective of deformation stage.

The luminescence peak of 420nm is attributed to exited Eu^{2+} ions which are created by recombination of free electrons from F-centers and Eu^{3+} ions.

The deformation luminescence takes place along slip bands and the most intense luminescence occurs in the intersection area of slip bands.

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Figure captions

Fig 1. Deformation luminescence observing system. A sample is deformed by compression. Luminescence is observed using a photomultiplier and the data are stored in a digital oscilloscope.

Fig.2. Absorption spectra of X-irradiated KCl:Eu^{2+} crystals.

Fig.3. Variations of stress(a) and luminescence(b) during plastic deformation and under intermittently superimposition of ultrasonic oscillation.

Fig.4. Relations of SL/DL and stress decrement due to superimposition of oscillation with strain.

Fig.5. Relation between stress decrement and SL/DL.

Fig.6. Variation of deformation luminescence spectra with strain.

Fig.7. Photograph of deformation luminescence.

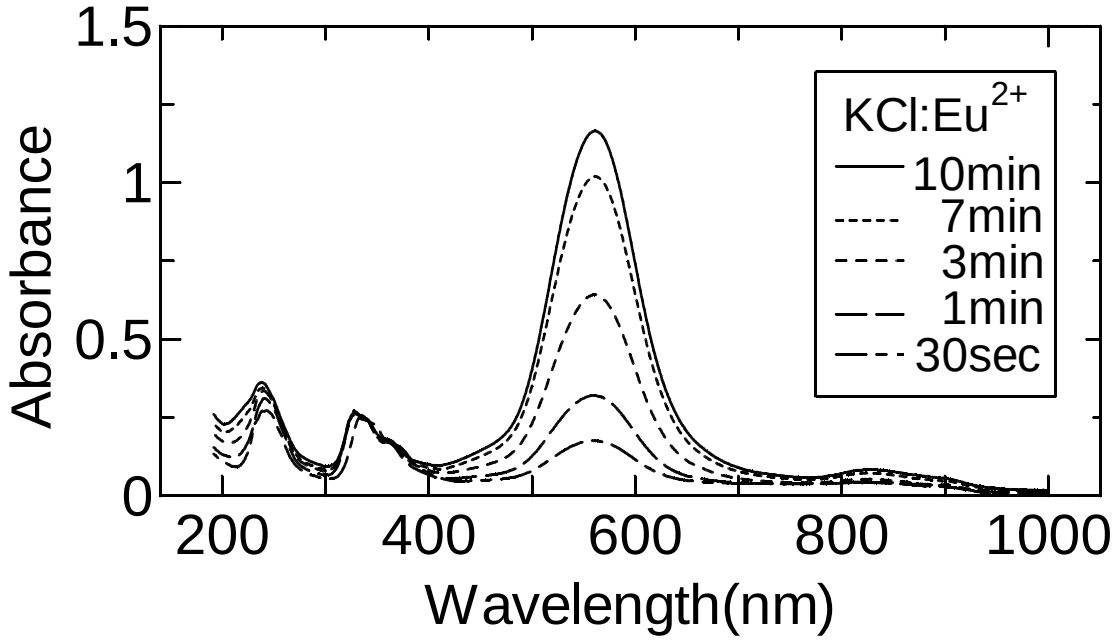

1.5
1
0.5
0
Absorbance
200
400
600
800
1000
Wavelength(nm)
KCl:Eu2+
10min
7min
3min
1min
30sec

Fig.2

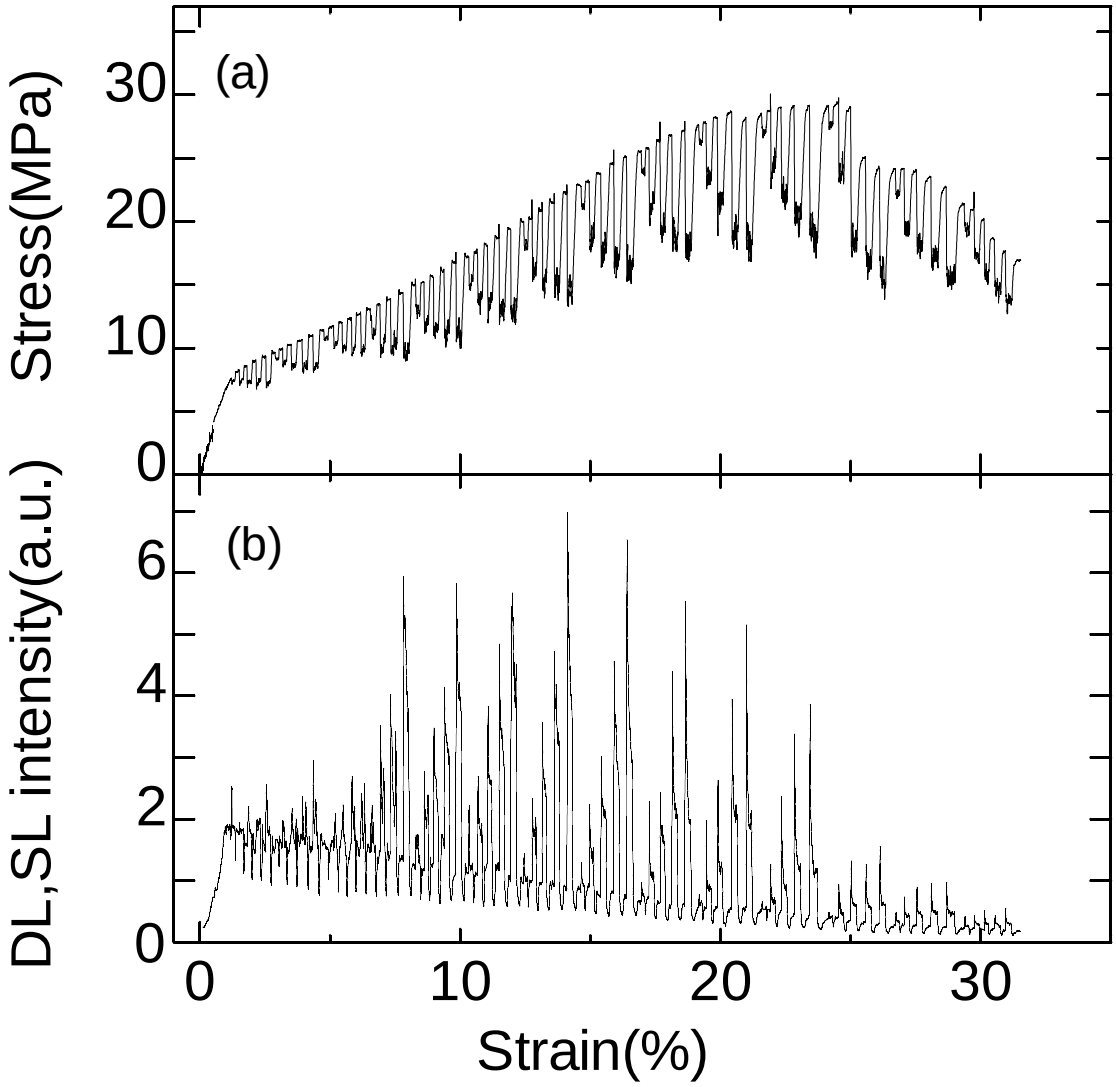


Fig.3

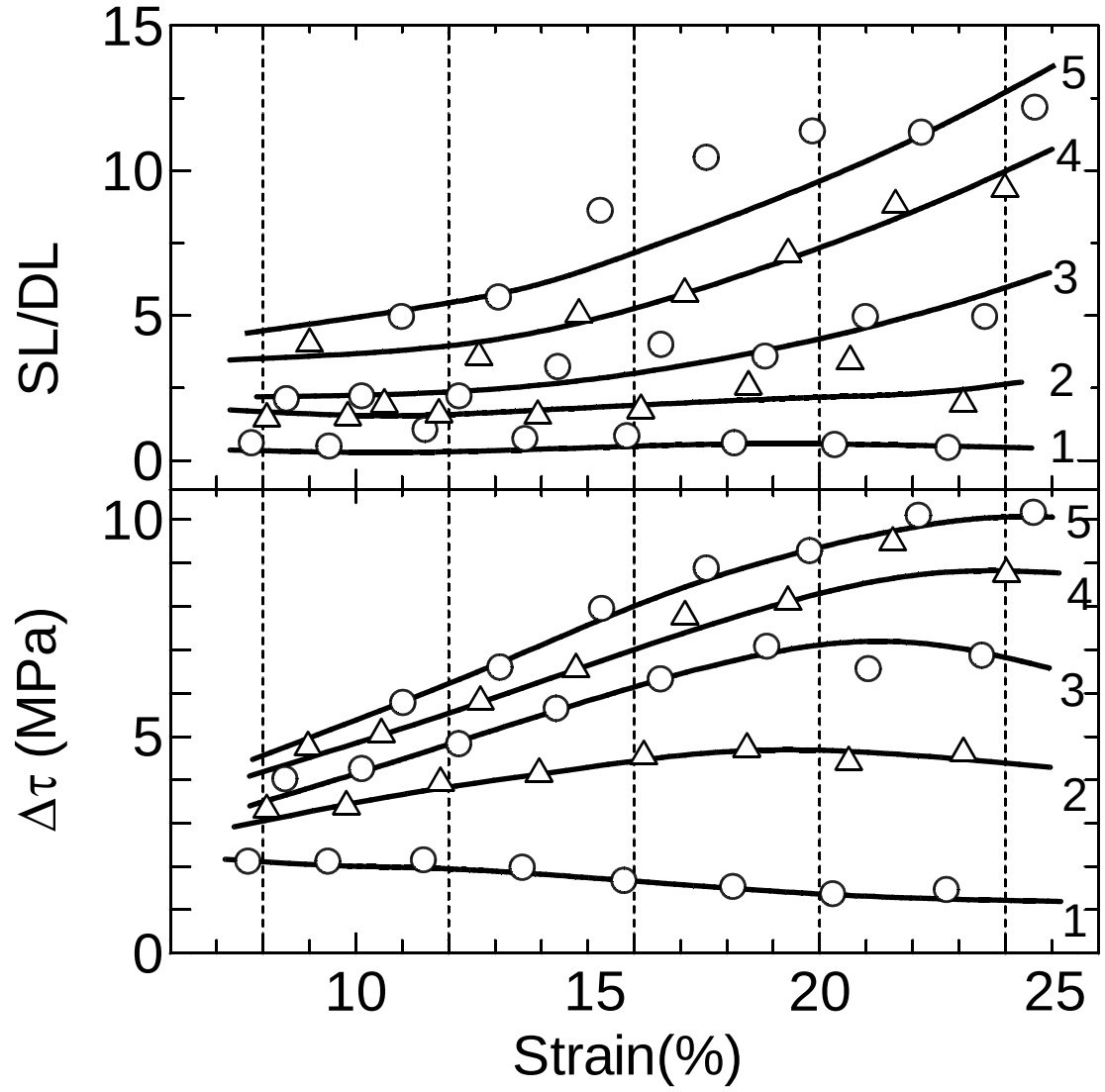


Fig.4

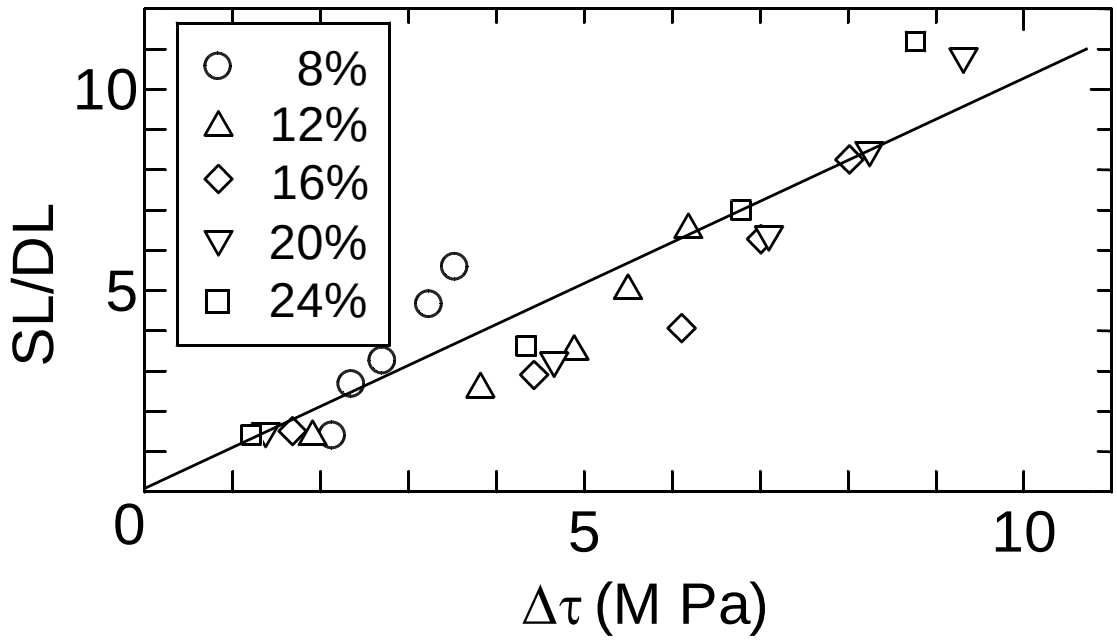


Fig.5

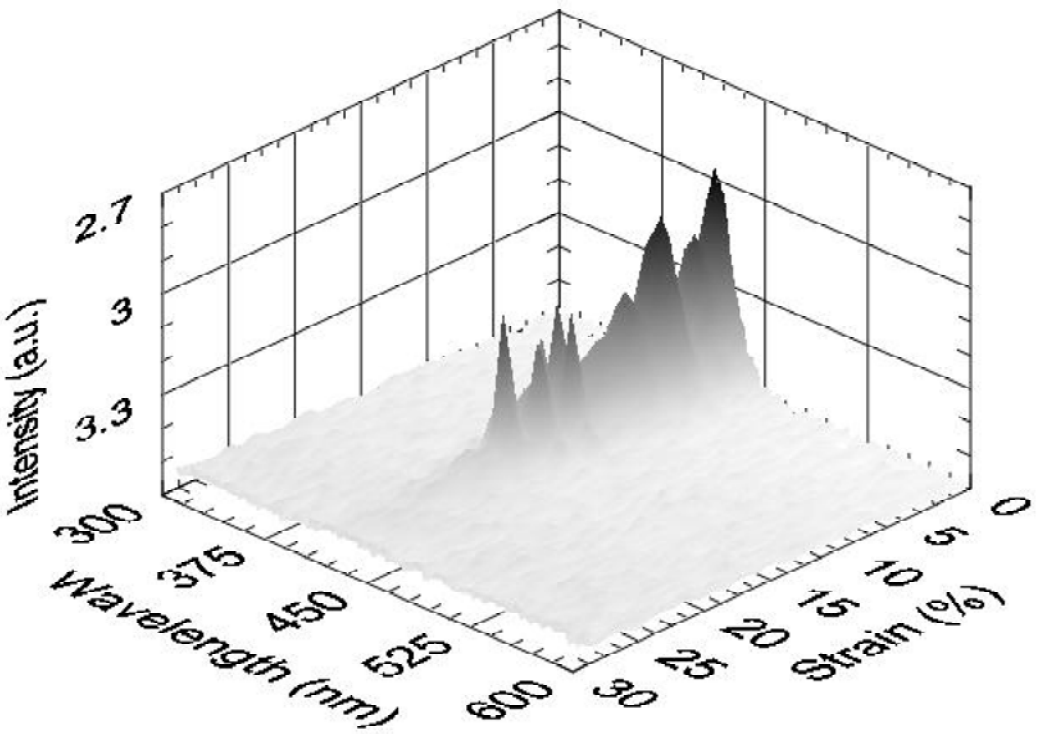
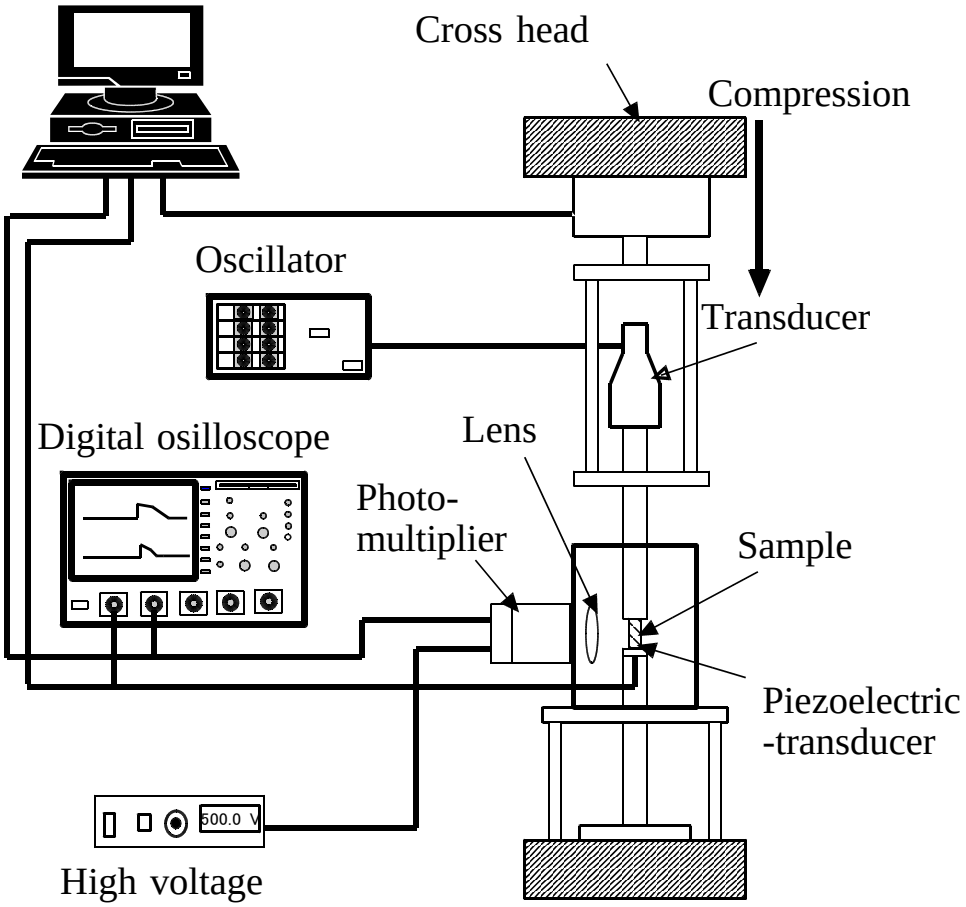
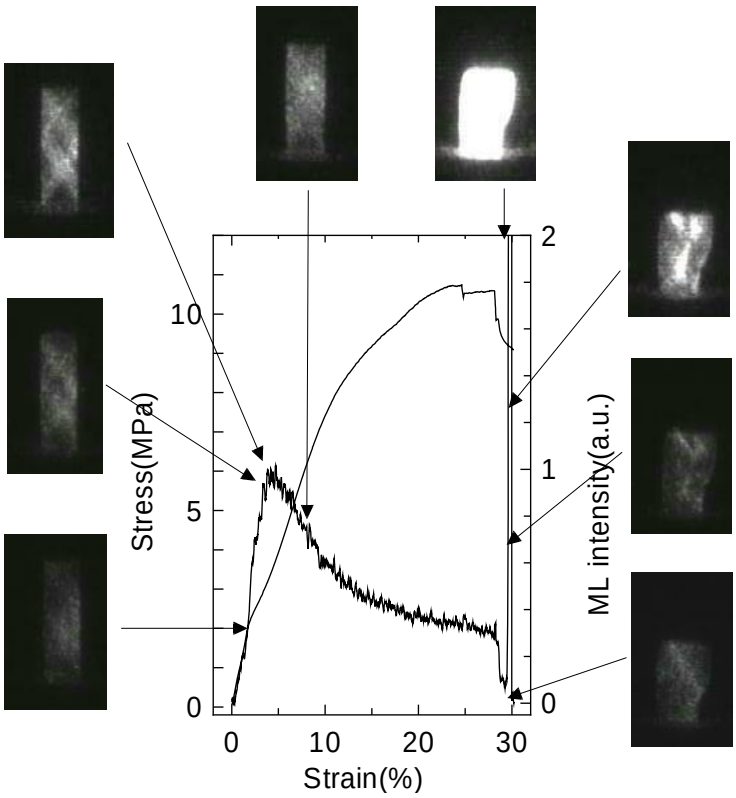

Intensity (a.u.)
2.7
3
3.3
300
375
450
525
600
Wavelength (nm)
0
5
10
15
20
25
30
Strain (%)

Fig.6

Fig.1


Cross head
Compression
Oscillator
Transducer
Digital osilloscope
Lens
Photo-
multiplier
Sample
Piezoelectric
-transducer
500.0 V
High voltage


Stress(MPa)
ML intensity(a.u.)
Strain(%)
0
5
10
0
1
2
0
10
20
30