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Ultrasonic Attenuation and Some Related Behaviors in Irradiated Alkali Halide Crystals

Touichi Okada and Tokuo Suita
School of Engineering, Osaka University
Osaka, Japan

Sodium chloride crystals subjected to gamma irradiation from zero to about 10^7 röentgen in total dose at room temperature have been examined during deformation by the following experimental techniques: (i) observation of the yield point on the stress strain relationships, (ii) observation of the ultrasonic attenuation changes as a function of the load, and observation of birefringence band. The yield stress of irradiated sodium chloride crystal in general increase with gamma dose for the following pretreated specimens: (1) as annealed in air, (2) after plastic deformation of about two per cent, (3) about two per cent deformation and then annealed in air. The yield stress *vs.* irradiation dose curve of the crystals annealed in air has maximum at the total dose of about 1.5×10^5r and minimum at about 10^6r. The stress strain relationships and the attenuation change *vs.* stress when the specimen is under load are similar for the early stages of plastic flow in annealed crystal, namely, near the stress where macroscopic yielding occurred, appreciable increase in the damping were observed; with further deformation the damping saturates at a certain value. The higher the irradiation dose, the smaller this saturation values were found to be. For irradiated crystals attenuation decreases by small amount just above the yield stress notwithstanding the onset of plastic deformation, and increases with the increasing load. Birefringence due to dislocations in glide bands of sodium chloride crystals clearly shows the initiation of the plastic flow in an unirradiated crystal as well as irradiated. A tentative model is proposed to explain the above experimental results.

Introduction

The ultrasonic pulse method, as a tool of studying dislocation dynamics in crystals has been considerably developed.[1),2)] The interaction between radiation-induced defects and dislocations can be conveniently studied with this technique when the specimens are subjected to irradiation.[3)] Truell[4)] has shown that the attenuation of deformed sodium chloride crystals decreases and velocity change increases when the crystals were subjected to gamma irradiation. From the view point of the study on radiation hardening, however, the applicability of the technique is limited to very low irradiation dose. On the other hand, measurement of yeild stress or critical resolved shear stress allows us to study radiation hardening in the wider range of irradiation dose.

Using pulse method at megacycle frequencies, it is possible to observe the internal friction while the specimen is being stressed in a testing machine.[5),6)] Consequently when irradiated specimens were investigated with the method which may be called an application of pulse method to higher irradiation dose in radiation hardening study, we will be able to evaluate some of the features of the radiation induced defects and their influence on dislocations. The present study has been designed so that yield stress and attenuation change of irradiated sodium chloride crystal can be compared to the similar data from unirradiated crystals from the view point of the dislocation pinning mechanism.

Experimental

The experiments were carried out with sodium chloride crystals purchased from the Harshaw Chemical Company and with Ca^{++} doped crystals made by Kyropoulos method in our laboratory. The specimens for yield stress measurement had (100) faces, and measured about $4\times4\times10\,mm^3$ in size. After cleavage from a large block the specimens were annealed for about four hours at 650°C to remove the internal stresses produced by the cleavage damage, and then cooled to room temperature over a period of one day. Cobalt-60 was used as the radiation source with dose rate of 2×10^5r/hr.

Stress strain curves were determined in

compression using a 'soft' machine designed so as to give axial loading and to record the strain of the specimens as a function of stress. Stress rate of about 25g/mm²/sec was used for the determination of yield stress. To avoid stress concentration at the top and bottom surfaces of the specimens which were sandwitched by the two thick brass plates, the contact surfaces were lubricated by silicon oil (Dow Corning 704).

Attenuation were measured by the pulse echo method with 5MC X-cut quartz crystals. To make simultaneous measurement of stress-strain curve and attenuation the specimen was arranged as shown in Fig. 1. The

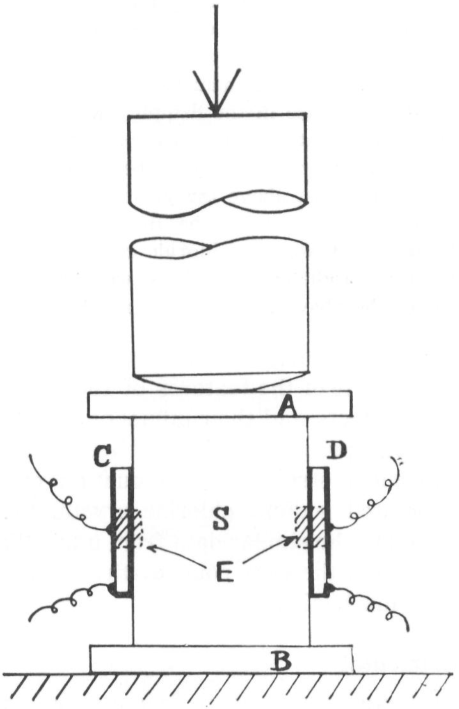


Fig. 1. Arrangement of the transducer and the receiver to the specimen. A, B; brass plates, C; 5 MC quartz transducer, D; 5 MC quartz receiver, E; binding agent, S; specimen.

specimen for attenuation measurement was sandwitched between two thick brass plates and both end surfaces, (100), of each specimen were lubricated by the Dow Corning Silicon oil. An X-cut quartz transducer and the same receiver was cemented to the specimen at the side portion, as shown in Fig. 1, and DC-200 were used as a binder at the contact layer between the quartz and the specimen. Under the condition mentioned above good pulse shape was obtained, though the background value of the attenuation is rather high.

All the measurements were made at room temperature in air throughout the experiment of this study.

Experimental Results

1 *Yield stress*

Fig. 2 shows the typical stress-strain curves

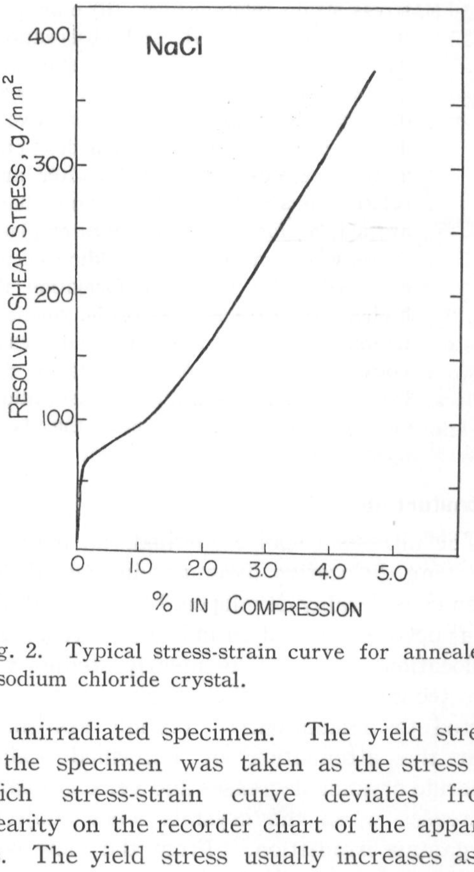


Fig. 2. Typical stress-strain curve for annealed sodium chloride crystal.

for unirradiated specimen. The yield stress of the specimen was taken as the stress at which stress-strain curve deviates from linearity on the recorder chart of the apparatus. The yield stress usually increases as a function of total gamma irradiation as shown in Fig. 3. The lower two curves shows the change in yield stress of annealed Harshaw sodium chloride crystal, while the upper curve represents the yield stress of the annealed crystals containing calcium chloride. It is important to notice here that both curves have maximum at about 10^5r and minimum at about 10^6r. The behavior does not seem to depend sensitively upon the calcium ion content for the specimen used in the present experiment. There is another experimental results that the purity of the sodium chloride crystal does not appreciably affect the behavior in thermal healing of gamma irradiation hardening.[7)]

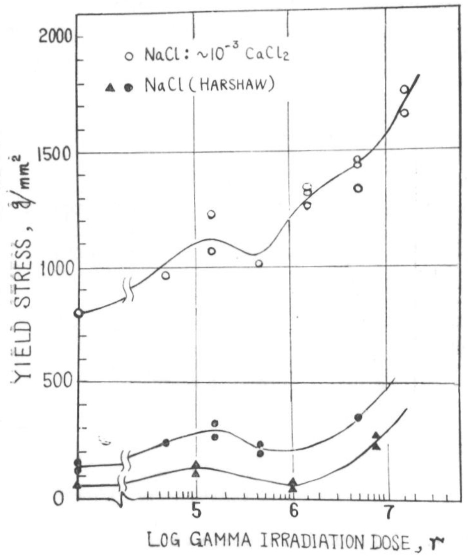


Fig. 3. Comparison of the yield stress glow curve as a function of gamma irradiation; the lower two curves indicate the results for different block from Harshaw sodium chloride crystal, while the upper curve shows the results for Ca^{++}-doped crystals.

In order to see whether the peak is related to fresh dislocation or not, we made the same irradiation hardening experiment with about two per cent deformed specimen. As shown in Fig. 4, no peak was seen in the relation,

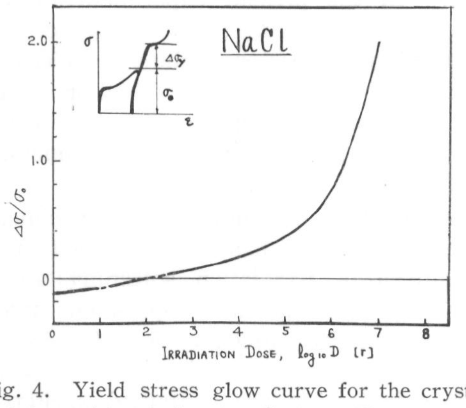


Fig. 4. Yield stress glow curve for the crystals deformed just before irradiation. Normalization was made as shown in the figure.

yield stress *vs.* irradiation dose. Because of the experimental difficulty the definite plastic strain (2%) could not be given to all the specimens, and a normalization was made represented by the ratio, the increment in the yield stress after irradiation divided by the shear stress given to the specimen before irradiation.

If the peak is related to some behavior of the dislocations which were annealed at high temperature, then we expect the amplification of this effect by introducing fresh dislocations and then annealing the specimens at high temperature. Fig. 5 represents the results of this series of experiment. The yield stress of these crystal shows monotonous increase as a function of the irradiation dose.

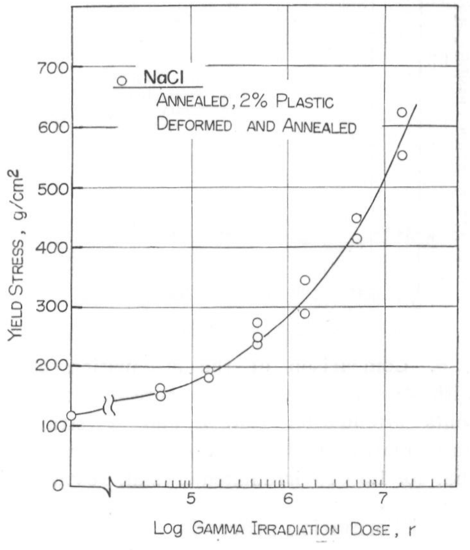


Fig. 5. Yield stress as a function of gamma irradiation show the results for plastic deformed up to about two per cent and annealed and again irradiated crystals.

2 *Ultrasonic attenuation due to plastic deformation*

Ultrasonic attenuation measurement made in sodium chloride during deformation will give information about the dislocation arrangement and its state in the crystal. If one does this kind of experiment on gamma irradiated crystals, it would be possible to obtain some knowledge about the radiation-induced point defects which may resist the motion of the dislocations. Moreover, if the temperature could be changed and the attenuation were measured at a definite temperature attenuation recovery could be investigated as a function of time. This sort of experiment leads to the study of the so-called "Köster Effect."

In the present experiment, however,

measurements were made only at room temperature, and recovery has not been investigated. Fig. 6 represents the attenuation-stress behavior and strain-stress relation in Harshaw sodium chloride crystal. As seen from the figure, for unirradiated crystal attenuation increases even the load is lower than the yield stress. Near the yield stress considerable increase in attenuation was observed and the attenuation increases as the

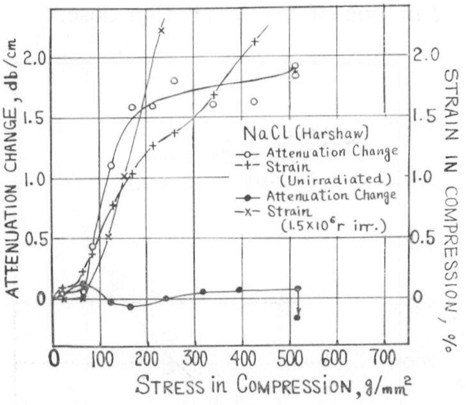


Fig. 6. Comparison of the attenuation change with strain, as the loading proceeds. ○ repersents attenuation change and + strain for unirradiated crystal, and ● and × represent that for 1.5×10^{6} irradiated crystal, respectively.

plastic deformation proceeds. The attenuation approaches to saturation value after all. Since the attenuation is proportional to dislocation density and the fourth power of loop length according to Granato and Lücke's theory, it is reasonable to assume that this phenomenon is related to the increase in dislocation loop length and also dislocation density. Qualitative as it is, there is rather distinct relationships between strain-stress curve and attenuation-stress curve for unirradiated crystals, *i. e.* large increase in attenuation coincides with the onset of plastic deformation. For irradiated crystals, however, it is not so. It is important to notice from the experimental view point that the attenuation measurement after the specimen is drastically deformed, say, more than two per cent is not reliable any more for the following reason. As the plastic deformation proceeds, the central part along the length of the specimen swells out in the direction through transducer and receiver crystals. Consequently, the increase in attenuation due to deformation may not be entirely due to dislocation damping but contain to some extent, partial failure in contact at the boundary layer between the piezo-electric crystal and the specimen.

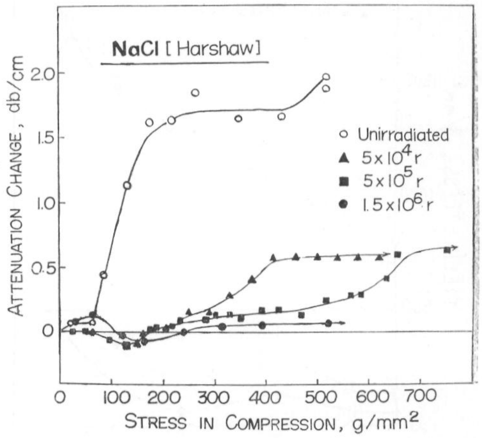
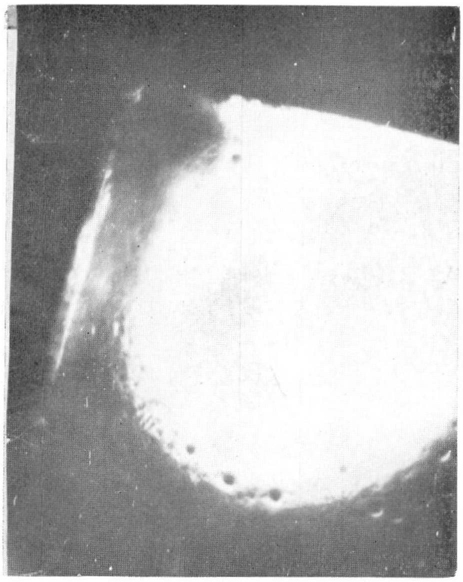


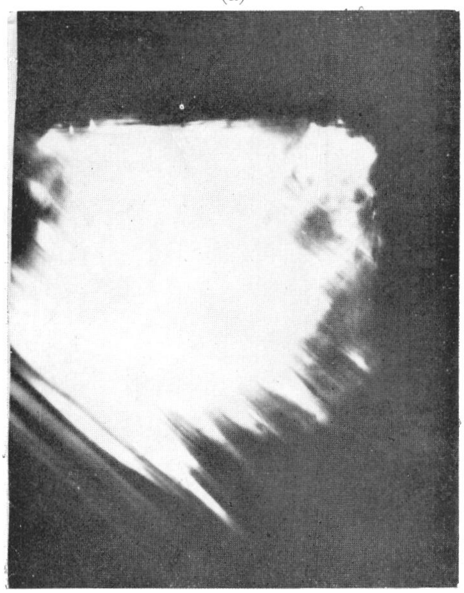
Fig. 7. Attenuation change as a function of the loading for various irradiation.

It is of importance to note in performing this sort of experiment for NaCl type crystal that orientation dependency of the attenuation of high frequency longitudinal acoustic wave is quite appreciable.[2] As has been shown, for the incident of longitudinal wave in the direction of Burgers vector of edge dislocation, attenuation is large, while in the direction perpendicular to the Burgers vector of edge dislocation the attenuation is comparatively small. In the present experiment, it was confirmed by observing the birefringence band that the Burgers vector of the slip dislocation has its components in the direction of the path of ultrasonic sound echoes. The conditions to have this seem to be the geometrical one (Photo. 1).

How about the case in irradiated specimens? Since our attention has been focussed to the phenomena of irradiation softening around $10^4\sim10^6$r, three annealed specimens which were gamma-irradiated by 5×10^4r, 5×10^5r, and 1.5×10^6r were tested. The results of these experiments are contained in Fig. 6. The behaviors of these crystals are quite complicated but interesting. At elastic range, the attenuation change was not seen for 5×10^4r and 5×10^5r gamma-irradiated specimens. The specimen which was irradiated by 1.5×10^6r has an increase in attenuation even when it is loaded below yield stress.



(a)



(b)

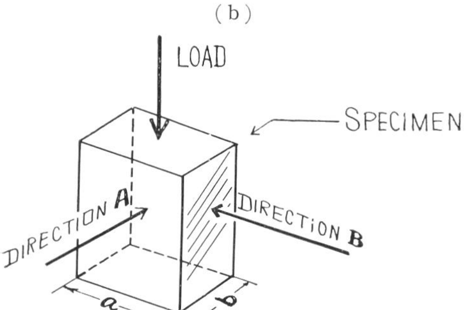


Photo. 1. Observation of birefringence bands by use of polarized light paralell to the direction A [Photo. 1(a)] and in direction B [Photo. 1(b)]. If a>b, slip usually takes place as shown in the figure. The attenuation were measured in the direction A.

Another interesting point is that the attenuation decreases by an amount just exceeding the experimental error at stress a little bit higher than the yield stress and again increases as the load is increased further. A saturation value of attenuation seems to decrease as the irradiation dose increases.

Discussion

Considering the previous experimental results, it is clear that any model which can account for the present results on gamma-irradiation hardening of sodium chloride crystal from the view point of dislocation pinning must satisfy the following conditions.

1) There must be at least two processes contributing to the dislocation pinning; one is eliminating the existing pinning points and the other creating the pinning agents. (Fig. 3).

2) The maximum and minimum appear only in the specimen which is annealed at high temperature and cooled slowly. Introduction of fresh dislocation due to plastic deformation of about two per cent, or annealing after plastic deformation "smooth out" this maximum and minimum.

3) For gamma-irradiated crystals the attenuation decreases by small amount when the specimen is compressed just above the yield stress.

To account for the phenomena described above, it should be necessary to consider two processes, one introducing hardening and the other introducing softening. Since this occurs only in annealed crystals but not in plastically deformed crystals before irradiation, one has to pay attention to the old dislocations in the crystal. Our qualitative model is as follows. As a hardening process we assume Gordon and Bauer's model.[8] Suppose a chlorine ion near a dislocation. Then a radiation ionizes the ion and makes it chlorine atom. This is more mobile, by virtue of its small size, moves to the dilatation side of the edge dislocation. Now we have chlorine ion vacancy and chlorine atom at the core of the dislocation. If each of these could then capture an electron, F center and chlorine jog are formed and this causes the dislocation pinning at low temperature. At higher temperature the F centers can diffuse beyond the recombination range. As a softening process we consider the following process. Suppose a locked dislocation mainly locked by multivalent impurity-vacancy pairs and chlorine ion ionized by radiation. Then through electronic process the electron thus produced relaxes the locking effect of multivalent impurity vacancy complex to the dislocation. We assume that this causes dislocation softening. This is very clude and there may be many points to be examined quantitatively. Since this occurred in a region surrounding the old dislocation core it may be called source hardening. It seems, however, that these two processes become comparable at an appropriate condition and cause maximum and minimum on the yield stress versus irradiation curve.

The results in Fig. 5. and Fig. 6 clearly show that the newly introduced dislocations by the plastic deformation after irradiation are more effectively pinned with increasing irradiation dose. Therefore, it is quite reasonable to suppose that the radiation induced defects are dispersed in the bulk of the lattice and make the dislocations hard to move with increasing gamma-irradiation dose. There may arise a question whether this is universal phenomena for all alkali halide crystals or unique one only seen in sodium chloride crystals related to some particular impurities. There also may be another problem how the kind of impurity in crystals affects the behavior described above. We have to await further systematic study to establish a definite model.

As the conclusion, we could say, as far as our experimental results concerns, that there seem two processes in radiation hardening; one contributes to softening and the other contributes to hardening.

Acknowledgements

Many thanks are due to Dr. N. Itoh and the staffs in Suita laboratory for helpful discussions concerning the hardening model, and to Dr. J. Mizuno and Mr. K. Morita of the Sumitomo Atomic Energy Institute for kindly performing gamma-irradiation. The authors are also indebted to Dr. W.G. Johnston for his stimulating discussions and suggestions.

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DISCUSSION

Crawford, J. H., Jr.: Work by K. Akimoto and W. A. Sibley at Oak Ridge may have some bearing on the radiation "softening" of alkali halides by irradiation. These workers study hardening of both Harshaw and Ca^{++} doped KCl by (a) quenching, (b) Co^{60} γ-ray and (c) 1.5 MeV electrons. Quenching to 0°C from successively higher temperatures causes hardening up to 350°C. Higher quench temperatures cause no additional hardening. This effect is interpreted as arising from the quenching-in of isolated cation vacancies and divalent cations. On γ-irradiation of quenched specimens, a decrease in flow stress is observed for exposures up to 10^7 R. For higher exposures additional hardening is observed. This observation suggests that the "softening" is caused by some radiolytic process involving either isolated cation vacancies or divalent cations.

Okada, T.: We want to clear out that point in future

Johnston, W. G.: The experimental observations are very interesting, particularly with regard to the maximum observed in the yield stress as a function of irradiation dose. Although the internal friction measuments show that dislocations are being pinned, I don't believe it will be fruitful to attempt to relate this pinning to the yield stress. It has been clearly shown in the past that the yield stress which is measured as described in the present experiments is a measure of the frictional stress encountered by fresh dislocation, and not of the state of pinning. Therefore the maximum in the flow stress will have to be explained in terms of the changes in frictional stress with irradiation.

Okada, T.: According to our definition of the source hardening we mean the increase in difficulty of the motion of as grown dislocation from the original position to certain distance due to some sort of locking agents located in the vicinity of the dislocation. There are of course some problems concerning the effective region of dislocation source, and defects introduced by irradiation might interact with the dislocation differently from in the outer region. Therefore internal friction measurement within the elastic region during plastic deformation may give some information about the motion of old dislocation in this region. Probably this effective region may be so small that the etch pit technique might not be able to observe the motion or behavior of the old dislocation there.

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Interstitial-Dislocation Interaction in Niobium*

D. H. BOONE AND C. A. WERT
University of Illionis, Urbana, Illinois, U. S. A.

Interstitial-dislocation interactions in niobium were investigated using the cold-work and Snoek internal friction peaks. Both the temperature of the cold-work peak and its height were found to depend on the amount of nitrogen removed from solution by dislocations in deformed specimens. Interstitial oxygen had only a minor effect and deformations beyond 5% were found to change the peak very little. The characteristics of the peak can be related to the interstitial content of the dislocation atmosphere and understood if these interstitials are assumed to interact to form clusters. These interstitial clusters relax in some cooperative manner which results in an order-of-magnitude increase in the relaxation strength per atom.

1. Introduction

Interstitial impurities, such as oxygen, nitrogen and carbon in b.c.c. metals are commonly thought to influence strongly the mechanical properties of these metals. This influence is thought to be particularly the result of interaction of the tetragonality of the strain field around the interstitial impurity with the shear strain fields of dislocations. Few definite measurements, however, have been able to show much about this interaction, which is believed to be much stronger than the hydrostatic interaction of substitutional impurities with dislocations. One of the types of measurement which has been thought to have promise in providing detailed understanding of the interstitial-dislocation complex is internal friction. The present paper reports such measurements for niobium.

The major effect used in this investigation is a damping peak found in deformed b.c.c. metals containing interstitials; we will call this the interstitial-cold-work peak. Such a peak in iron has been known for many years; it was first seen by Snoek, and later studied by numerous other investigators. This interstitial-cold-work peak exists in deformed b.c.c. metals in addition to the normal stress-induced-ordering peak also found in these alloys.[1)-3)] (This latter peak was also discovered by Snoek

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