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INTERNAL FRICTION OF Fe-B ALLOYS NEUTRON IRRADIATED AT LOW TEMPERATURE

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Measurements were made on the internal friction of Fe-B alloys irradiated by neutron at 16 °K to the dose of 3×10^{16} nvt (>1 MeV) and 6×10^{17} nvt (thermal). Boron was used to enhance the production of defects by the nuclear transformation $B^{10}(n,\alpha)Li^{7}$. Relaxation peaks were found in specimens containing dispersed fine precipitates of NbB_2 in range of B 500~7200 wt ppm and Nb 2000~30000 wt ppm. The most prominent peak is the one with the peak temperature of 169 °K at the frequency of 264 c/sec. Activation energy determined from the peak shift is 0.28 ± 0.01 eV, which is nearly equal to that of migration of self-interstitial reported on pure iron. However activation energy of the decay of peaks by annealing is about 0.7 eV. Interpretation was presented that the peak may be attributed to re-orientation of self-interstitials loosely bound to a boron atom.

1. Introduction

Irradiation of high energetic particles, neutron or electron, at low temperature provides a useful mean for fundamental studies of lattice defects in metals, since irradiation produces simplest types of lattice defects, self-interstitial and vacancy, and they are retained in as irradiated state at very low temperatures. So many works have been made on FCC metals [1], and recently on BCC metals [2]. Systematic studies for iron made by Grenoble group [3][4][5] provided clear interpretations for characteristics of self-interstitial, *i.e.* its orientation in the lattice, re-orientation and migration energies etc., based on numerous data of measurements on electric resistivity, internal friction, specific heat and magnetic after-effects etc.

However, it is considerd to be worth-while to re-examine these results, because definite conclusions are yet in a state of controversies [1], and sometimes liable to the effects of impurities particularly in BCC metals [2].

This paper is an trial to measurements of self-interstitials in iron by means of internal friction [7][8]. We used Fe-B alloys because available dose of neutron was found to be too low to detect relaxation peaks in pure ion, and the production of defects can be enhanced by nuclear transformation associated with B.

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2. Experimental Procedure

2.1. Instruments

Low temperature neutron irradiation was made using the low temperature loop of Kyoto University Reactor, (LTL of KUR). The main characteristics of the loop are as follows. The loop is horizontal type one circulating helium gas cooled by cryogenic engine with cooling power of 30 W at liquid helium temperature, and is capable to maintain the specimens at 16 °K for 80 hr when the output power of the Reactor is 5 MW. The neutron flux at the top of the loop is $\phi_f=2.98\times10^{10}$ n/cm^2 sec MW for fast neutron (>1 MeV), and $\phi_{th}=4.62\times10^{11}$ n/cm^2 sec MW for thermal neutron so that the total dose is $\Phi_f=3.75\times10^{16}$ n/cm^2 (>1 MeV) and $\Phi_{th}=5.90\times10^{17}$ n/cm^2 for 70 hr irradiation at 5 MW.

Measurements of irradiated specimen can be made by two ways, the one is in-loop measurements for internal friction and electric resistance together with temperature of the specimen. On the other hand, specimens can be taken out from the loop and dropped into a liquid helium cryostat without increasing the temperature of specimens after irradiation. Various out-loop measurements are then made on these specimens. Details of the loop and the apparatus of in-loop measurement of internal friction were explained in the references [6)7)8)].

2.2 Specimens

Since we found that available fast neutron dose is too low to detect stress relaxation peak due to self-interstitials in internal friction of pure iron, we doped boron to pure iron with intent to increase the number of defects using the nuclear reaction $B^{10}(n,\alpha)Li^7$.

At first, boron of 200~10000 ppm was doped into pure iron by melting in electron beam melting furnace, then the ingot is formed to plates by cold work, and heated to 1150 °C for 1 hr and quenched to room temperature. By these procedures, however, solid solutional B was found to be about 200 ppm and remainder of boron was precipitated in the form of large grain of Fe_2B. So that we further added Nb with intent to obtain fine dispersed precipitates of NbB_2 in the specimens [9)].

Nb and B, or 99.5 % B^{10} in some cases, were added to pure iron in the electron beam melter, then the ingot was zone-melted by 1 pass in the zone-melting furnace with high frequency induction coil. The ingot was formed to plates of 1×7×100 mm by cold work, then annealed at 1200 C° for 1 hr and 800 °C for 2 hr and cooled in furnace. Precipitates of NbB_2 were observed by scanning type X-ray micro-analyser. Precipitates of NbB_2 with size of about 1 μ are found to be dispersed with average distance of about 10 μ. We prepared 0.2×2×100 mm foils of pure iron and Fe-boron alloys for in-loop measurements of electric resistance, and specimens of internal friction listed in Table 1.

Table 1. List of specimens for measurements of internal friction. A.C.; air cooling, F.C.; furnace cooling, W.Q.; water quench, f; resonant frequency at room temperature.

Spec. No.	Date	Nb (wtppm)	B (wtppm)	Heat Treatments	Size	f c/sec	Irrad.
1	1969. 7.10	0	10000	700°C(1hr)→1150°C(1hr)→A.C.	5.068×0.688×90.10	486	1MW 30hr
2	7.11	0	200	〃 1150°C(2hr)→A.C.	5.192×1.134×90.45	727	〃
3	9.13	3300	800	1200°C(1hr)→A.C. 800°C(2hr)F.C.	5×0.943×80	801	5MW 70hr
4	〃	10000	2400	〃 〃	5×0.793×80	667	〃
5	〃	30000	7200	〃 〃	5×0.861×80	720	〃
6	〃	10000	2400	〃 〃	5×0.855×80	715	〃
7	〃	3300	800	〃 〃	5×1.063×80	905	〃
8	11.	3300	800	〃 〃	~5×~0.6×100	288	〃
9	〃	0	200	〃 〃	~5×~0.6×100	290	〃
10	〃	3300	800	〃 〃	~5×~0.6×100	264	〃
11	〃	3300	800	〃 〃	~5×~0.6×100	280	〃
12	〃	3300	800	〃 〃	~5×~0.6×50	2088	〃
13	1970. 3.20	3300	800	1200°C(1hr)W.Q. 880°CAr(10hr)F.C.	7.9×0.51×100	267	〃
14	3.20	3300	800	〃 〃	8×0.48×100	257	〃
15	1969. 7.	0	Fe_2B	900°C(24hr) in B powder	~5×~1×95	730	1MW 30hr
16	7.	0	0	700°C(1hr)→850°C(24hr)→F.C.	4.86×1.11×9.03	733	〃
17	7.17	0	10000	700°C(1 hr)1150°C(1hr)→A.C.	5×0.5×54	1064	〃
18	1970, 10.	2000	B^{10}500	1200°C(1hr)→A.C., 800°C(2hr)F.C.	5×0.7×50	2065	5MW 70hr
19	〃	〃	〃	〃	5×1×90	715	〃

3. Experimental results

3.1. In-loop measurements of electric resistance

Increase of electric resistance of pure iron and 500 ppm B^{10}-Nb-Fe during irradiation is shown in Fig. 1, and the isochronal annealing curve of pure iron in Fig. 2 which was made by out-loop measurements.

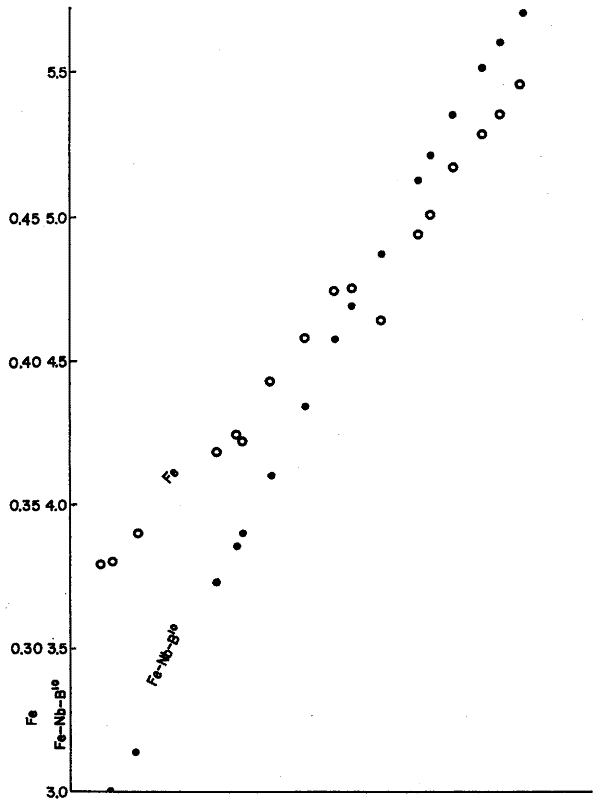

Fig. 1 Increase of electric resistance of pure iron and 500 ppm B^{10}-2000 ppm Nb-Fe during irradiation of 70 hr. Abscissa is time.

3.2. In-loop measurements of internal friction

Fig. 3 shows the change of 10000 ppm B-Fe specimen with decreasing of temperature before irradiatoin (curve(b)) and increasing of temperature after irradiation of 30 hr in 1 MW (curve(a)). In this case, however, no peak was detected because precipitates of Fe_2B had grown to too large sizes.

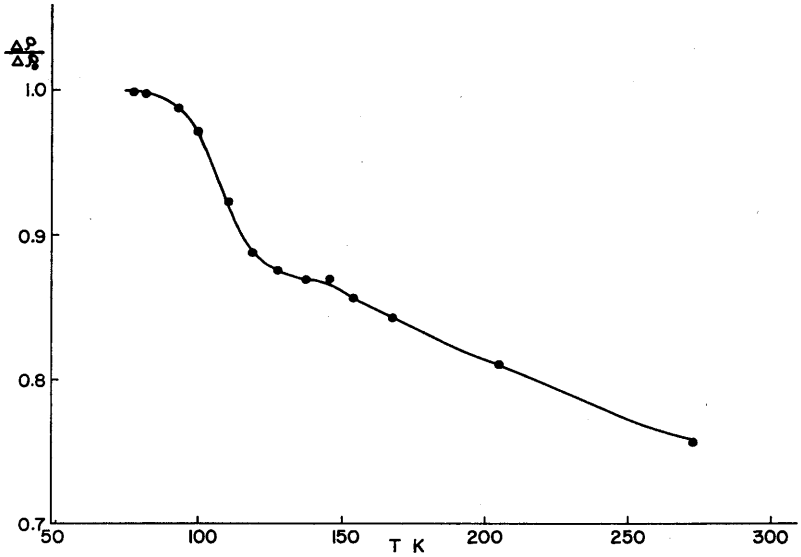

Fig. 2 Isochronal annealing curve of electric resistance of pure iron in Fig. 1. Annealing time is 10 min.

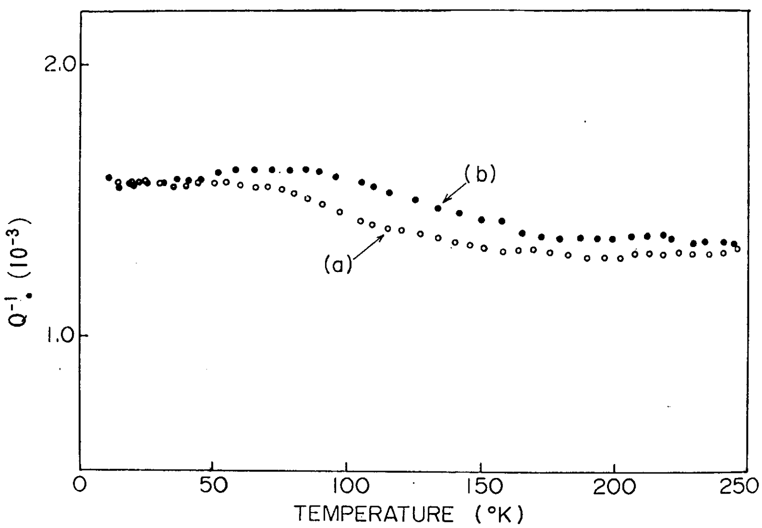

Fig. 3 In-loop measurements of 10000 ppm B-Fe. a); after, b); before irradiation, specimen No. 17.

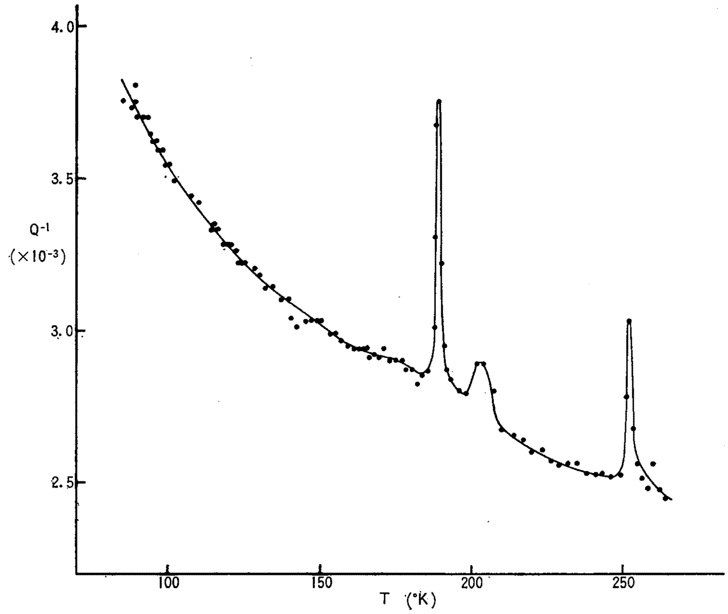

Fig. 4 Internal friction of 200 ppm B-Fe, specimen No. 9, out-loop measurement.

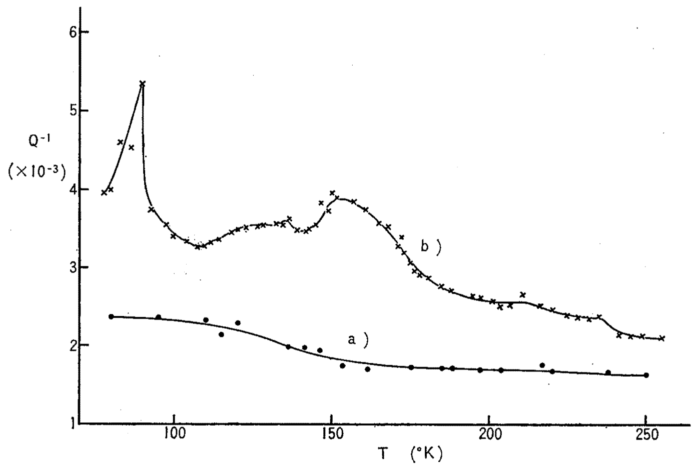

Fig. 5 Internal friction of 10000 ppm B-Fe, specimen No. 1. b) is the first run, and a) the second run.

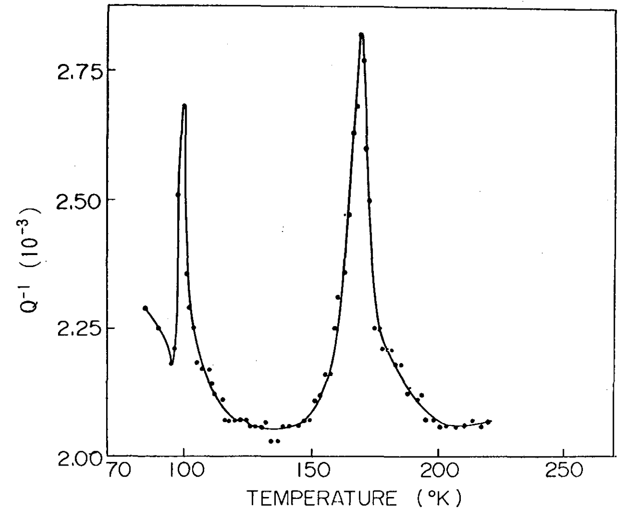

Fig. 6 Internal friction of 800 ppm B-Nb-Fe, specimen No. 10, f=264 c/sec.

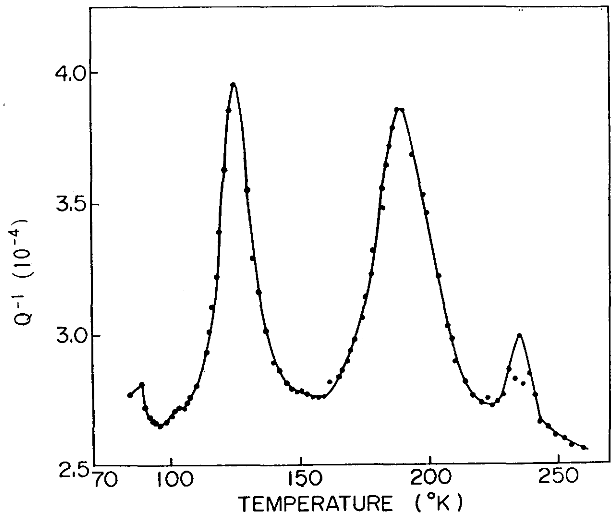

Fig. 7 Internal friction of 800 ppm B-Nb-Fe, specimen No. 12, f=2060 c/sec.

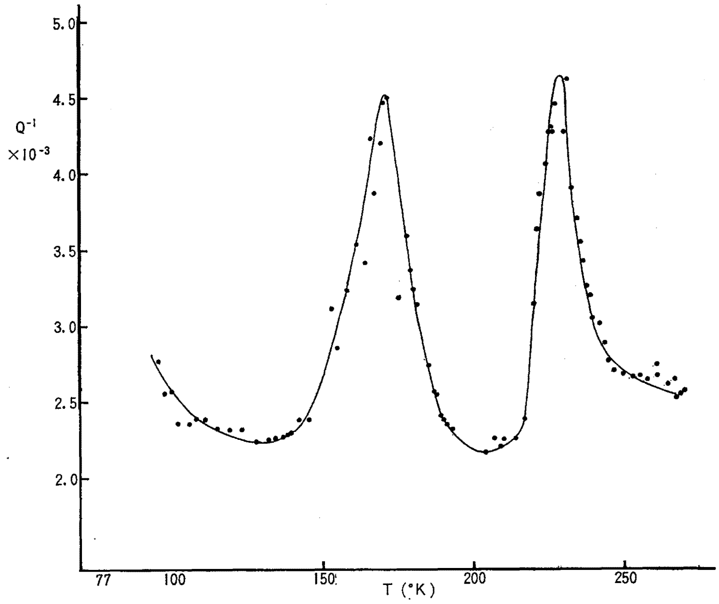

Fig. 8 Internal friction of 800 ppm B-Nb-Fe, specimen No. 13.

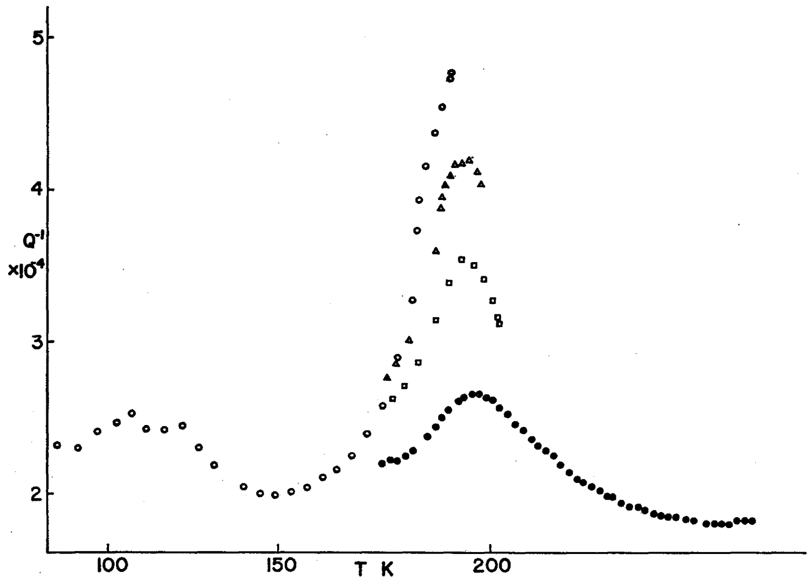

Fig. 9 Internal friction and its decay of 500 ppm B^{10}-Nb-Fe, specimen No. 18.

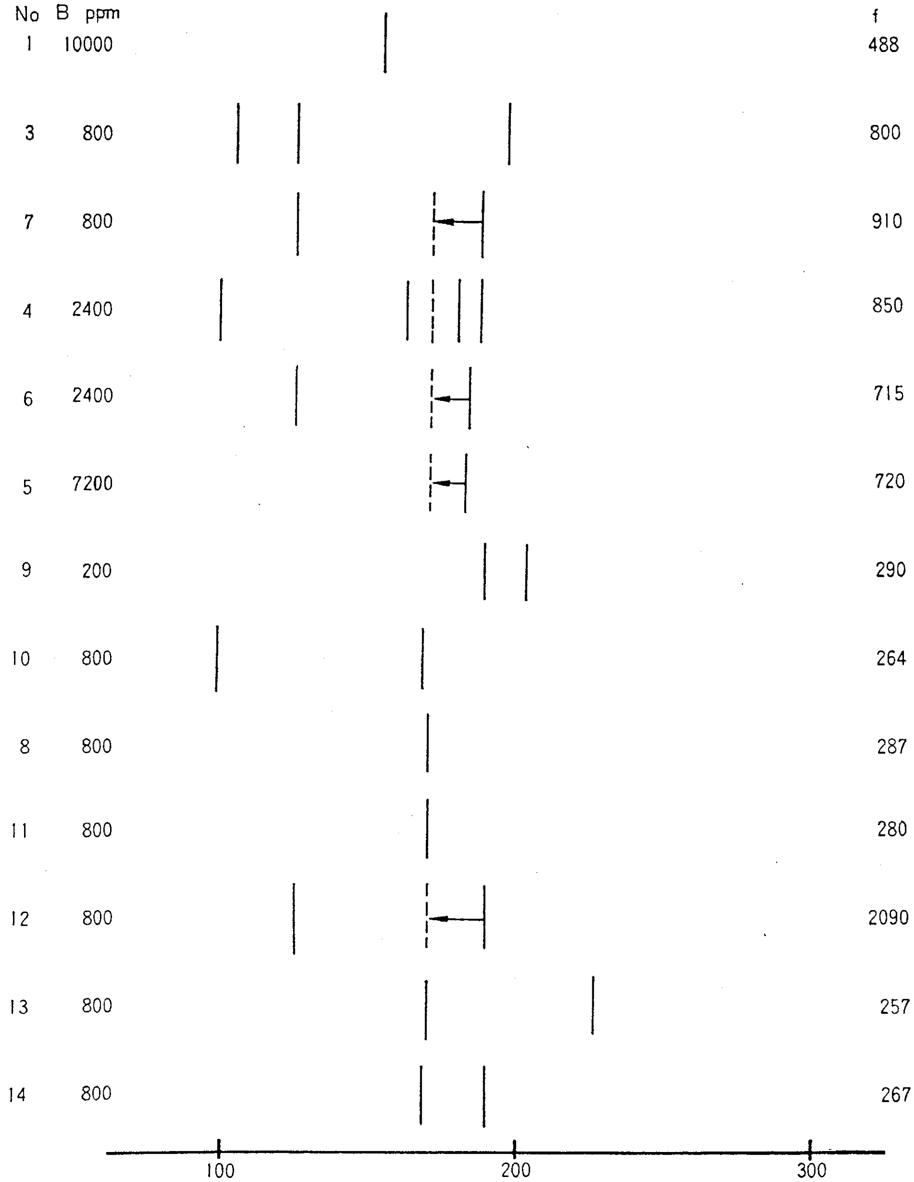

Table. 2 Temperatures of peaks observed in specimens with various frequencies. Dotted lines are those corresponding to the frequency of 264 c/sec calculated by the equation (1) with activation energy of 0.28 eV.

Table 3. Activation energies of relaxation peaks determined by the peak shifts in specimens No. 10 and No. 12.

Specimen	Peak 1		Peak 2	
	T_{p1} (°K)	f (c/sec)	T_{p2} (°K)	f (c/sec)
No. 10	100	265	169	264
No. 12	125	2098	190	2086
Act. energy (eV)	0.09		0.28	

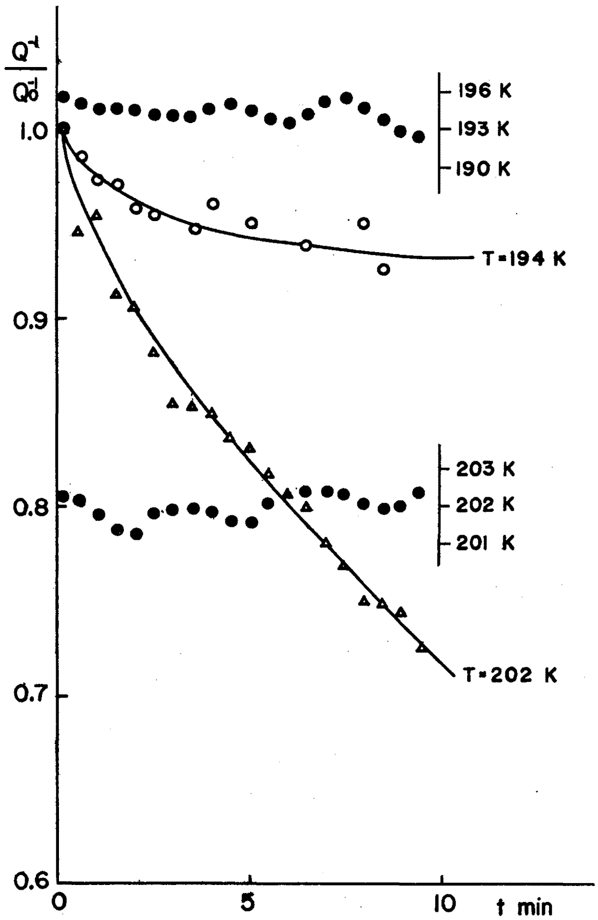

Fig. 10 Decay of internal friction peaks at peak temperatures on 500 ppm B^{10}-Nb-Fe, specimen No. 18 and No. 19.

3. 3. Out-loop measurements of internal friction

On Fe–B alloys peaks were detected only in the specimens of 200 ppm B–Fe irradiated for 80 hr by 5 MW, Fig. 4, and 10000 ppm B–Fe 30 hr 3 MW, Fig. 5. On the contrary, peaks were always detected on specimens of Nb–B–Fe alloys. Some examples are shown in Figs. 6, 7, 8. The temperatures of peaks are classified into three groups, *i.e.* the group near 100 °K, near 170 °K and higher than 200 °K, as shown in Table 2. On the groups near 100 °K and higher than 200 °K, reproducibility of peaks was not so good. Peaks near 100 °K were not found on specimens placed for long time at liquid nitrogen temperature. Activation energies were calculated from the peak shifts, *i.e.* the differences of peak temperatures between the specimen which have frequency of 264 c/sec, specimen 10, and that of 2090 c/sec, specimen 12, and listed in Table 3. Recovery of peaks were then measured on the specimens of No. 18 and No. 19. Fig. 9 shows the decay of peak by annealing at the temperature of peak temperature of 202 °K on the specimen No. 18 which has the length 50 mm and f=2065 c/sec. Similar measurements were made on specimen No. 19 which has the peak temperature of 194 °K, f=715 c/sec and the length of 90 mm. The time changes of decays of the peaks in the two specimens are shown in the Fig. 10, where internal friction Q^{-1} is normalized by $Q^{-1}_{t=0}$.

4. Discussion

According to Dautreppe [4] the neutron dose to produce detectable relaxation peak of $Q^{-1}\sim 10^{-3}$ is about 2×10^{17} nvt which is one order higher than the present dose of 3.7×10^{16} nvt. So that we doped B in order to enhance the production of defects. The isotope B^{10} contained by 18.8 % in natural boron induce the nuclear transmutation B^{10} (n, α) Li^7 by absorbing thermal neutron, and the energetic particles Li^7 and α, which have mean kinetic energies of 0.84 MeV and 1.47 MeV respectively, produce defects within the range of mean free path of about 1 μ. Absorbing cross section of B^{10} in the nuclear transmutation is $\sigma_{th}=4010$ barn, which is very large compared with the scattering cross section in collision of fast neutron with Fe atoms $\sigma_f \simeq 4$ barn. On the other hand, according to the measurements of electric resistance, the increase of number of defects in the specimen containing 500 ppm B^{10} is estimated to be about 20 times that of pure iron. The increase of the number of defects produced the peak of about $Q^{-1}\sim 4\times 10^{-3}$ in the present experiment. The results is consistent with Dautreppe's experiments, yet the reason is not clear.

The peaks near 170 °K have good reproducibility as shown in Table 2, where equivalent peak temperatures corresponding to the frequency of $f=264$ c/sec were calculated using the activation energy of 0.28 eV in Table 3, and listed in Table 2 by dotted lines. The peak temperature T_m is related to resonant frequency f by

$$f = f_0 \exp -H/kT_m , \tag{1}$$

where H is activation energy and have the value of 0.28 ± 0.01 eV, and f_0 is the frequency factor having the value 8.8×10^{10} sec^{-1}. The equation predicts that $T_m = 126$ °K for $f = 1$ c/sec. These values are nearly equal to those obtained by Dautreppe[4], 0.26 ± 0.03 eV, 124 ± 2 °K, $f = 2.3$ c/sec, and those obtained by Bilger[3)5], 0.25 ± 0.03 ev, 126 °K, $f \sim 1$ c/sec. They attributed the peaks to free migration of self-interstitials.

However, annealing experiment Fig. 10 shows the activation energy of $0.6 \sim 0.7$ eV for the stage of $4 \sim 6$ % annealing of $Q^{-1}/Q^{-1}_{t=0}$ contrary to the case of Bilger[3] where the activation energy of annealing coincided with that of re-orientation. The difference of activation energies of relaxiation and annealing means that self-interstitial is rotating around some trap center with the re-orientation energy of 0.28 eV and the binding energy of 0.7 eV.

Since the re-orientation energy is not so much affected by the trapping contrary to ordinary cases[2], we may suppose a loose trapping peculiar to present specimen. B may be a possible candidate, since substitutional B[9] which have smaller atomic size than that of iron is considered to provide a weak trapping center for interstitial atom.

We may point out further the fact that free interstitials could not be detected by the present method of lateral vibration, because the frequency used is larger than the jump number of annihilation of interstitial, which was reported as about 200 according to Bilger[3], consequently interstitials could not survive so long time at high temperature of 170 °K as required for present measurements.

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