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Effects of 800 MeV Xe Irradiation on 2H-NbSe₂ Single Crystals

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Abstract. Effects of artificial defects on 2H-NbSe₂ single crystals introduced by 800 MeV Xe irradiation with a dose equivalent matching field B_{Φ} up to 8 T were studied. The bulk magnetization measurements performed by a commercial SQUID magnetometer reveal strong enhancement of critical current density in NbSe2 after introducing defects by 800 MeV Xe irradiation. $T_{\rm c}$ is found to be robust against the 800 MeV Xe irradiation, while c-axis lattice parameter shows an obvious increase with increasing B_{Φ} .

1. Introduction

Enhancement of critical current density (J_c) is one of the challenges for the practical application of superconductors. The energy dissipation in superconductors caused by the motion of vortices has long been a big obstacle to obtain superconductors with high J_c [1]. Introduction of artificial defects has been proved to be an effective way to suppress vortex motion and enhance the J_c [2, 3]. In general, there are two ways to introduce artificial defects in superconductors, which are chemical and physical methods. However, in the case of chemical methods, not only the defects are introduced into the superconductors but also other unavoidable reactions can occur.

To study the effects of the artificial defects on superconductors, the method introducing defects has to be capable of introducing defects effectively without destroying the sample. Introduction of defects by heavy-ion irradiations has been proved to be an effective way with little damage on the bulk of the sample [4-7]. Through heavy-ion irradiations, columnar defects have been successfully introduced in cuprate [8] and iron-based [9, 10] superconductors, and the enhancement of J_c has been confirmed in those high-temperature superconductors. While there have been many reports on the effects of artificial defects in cuprate and iron-based superconductors with high anisotropy and Ginzburg numbers, very limited number of studies have been conducted on conventional superconductors with small anisotropy and Ginzburg numbers. 2H-NbSe2 is one of the representative conventional superconductors with a layered structure, the transition temperature $T_c \sim 7$ K, and Ginzburg number ~8×10⁻⁷ [11]. It is also well known that the NbSe₂ pristine crystal is a very clean system with a small residual resistivity, which is a good candidate to study the effects of artificial defects introduced by heavy-ion irradiations. In the present study, we investigated the effects of 800 MeV Xe irradiation on physical properties of 2*H*-NbSe₂ single crystals.

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2. Experimental details

2H-NbSe₂ single crystals were prepared by iodine vapor transport method [12]. We used high pure Nb powder (3N), Se shot (5N) and I_2 shot (5N) as starting materials. Stoichiometric ratio of Nb and Se with total weight of ~ 2 g were sealed in an evacuated quartz tube together with transport agent of I₂. Roughly 2.5 mg/cc of iodine were used as transport agent. The quartz tube was put in a tubular furnace with one end close to the center (high temperature part) and another end ~16 cm from the center. Then it was slowly heated up to $\sim 800^{\circ}$ C and held at that temperature for two weeks. 800 MeV Xe irradiation was performed at NIRS-HIMAC in Chiba. Before performing the irradiation, NbSe₂ single crystals were cleaved to thin plates with a thickness $\sim 15 \,\mu\text{m}$ along the *c*-axis, which is less than half of the projected range for 800 MeV Xe irradiation in NbSe₂ of \sim 35 µm calculated by SRIM-2008 (the Stopping and Range of Ions in Matter-2008) [13]. 800 MeV Xe ions were irradiated along the crystallographic *c*-axis of the crystal up to dose-equivalent matching field of $B_{\Phi} = 8$ T. Changes in the lattice parameter induced by the irradiation were characterized by X-ray diffraction (XRD) measurements using a commercial diffractometer (Smartlab, Rigaku) with Cu Ka radiation. Magnetization measurements were performed using a commercial SQUID magnetometer (MPMS-XL5, Quantum Design). After measuring the magnetic hysteresis loops (MHLs), we use the extended Bean critical model [14] to estimate the J_c . In this model, J_c [A/cm²] is given by

$$J_{\rm c} = \frac{20\Delta M}{a(1-a/3b)} \quad (b > a),\tag{1}$$

where ΔM [emu/cc] is the difference of magnetization when sweeping the external field up and down. a [cm] and b [cm] are sample width and length, respectively.

3. Results and discussion

As shown in the upper inset of figure. 1, the obtained crystals have well-defined facet with dimensions up to ~5 × 5 × 0.1 mm³. The temperature dependence of resistivity for a pristine NbSe₂ is shown in figure. 1. The estimated residual resistivity ratio (RRR = ρ (T = 300 K)/ ρ (T = 8 K)) is ~50 with a sharp superconducting transition at ~7.16 K, defined by the onset of resistivity drop, as shown in the lower inset of figure. 1. These data indicate the quality of the NbSe₂ crystal is very high. Figures 2(a) and (b) show the evolution (008) peaks of the XRD patterns, and the *c*-axis parameter as a function of B_{Φ} , respectively. The two peaks in figure. 2(a) originate from K α_1 and K α_2 X-ray beams. The *c*-axis lattice parameters were calculated based on the XRD data for (002), (004), (006), and (008) peaks. As shown in figure. 2(b), the *c*-axis lattice parameter increases linearly as a function of B_{Φ} after the 800 MeV Xe irradiation. Possible reason for the expansion along the *c*-axis after the irradiation can due to the strain from the expansion of amorphous linear tracks.



Figure 1. ρ -*T* curve of 2*H*-NbSe₂ single crystal synthesized by the iodine vapor transport method. The upper inset is a photo of NbSe₂ single crystal with well-defined facet, while the lower inset is the blow-up of ρ -*T* curve near the transition temperature.



Figure 2. (a) X-ray diffraction patterns of (008) peaks in NbSe₂ single crystals before and after 800 MeV Xe irradiation up to $B_{\Phi} = 8$ T. (b) *c*-axis lattice parameter as a function of B_{Φ} for NbSe₂ irradiated by 800 MeV Xe. (c) Temperature dependence of the normalized magnetization M(T) / M(5 K) at H = 5 Oe for pristine and 800 MeV Xe irradiated NbSe₂ single crystals with $B_{\Phi} = 1$ T, 2 T, 4 T, and 8 T.

Magnetization as a function of temperature (*M*-*T* curves) of NbSe₂ before and after irradiation by 800 MeV Xe with different B_{Φ} are shown in figure. 2(c). Obviously, T_c does not change within the accuracy of the measurement. This is consistent with the Anderson theorem which predicts no effect on T_c by point disorders in isotropic superconductors [15]. It is in good contrast with the effect of 800 MeV Xe irradiation on iron-based superconductors, where suppression of T_c by ~7.5% at $B_{\Phi} = 10$ T is observed [9]. In addition, it contradicts with the effect of electron irradiation on NbSe₂ reported by Cho *et al.* [16], where they reported ~8.3% change of T_c after irradiation of electrons by 2.4 C/cm². However, it should be noticed that the defect density at $B_{\Phi} = 10$ T of 5×10^{11} cm⁻² is much smaller than the dose of electron irradiation. Alternatively, T_c can be affected by the change in the lattice parameter through pressure dependence of T_c . Pressure dependence of T_c and the ratio of *c*-axis lattice parameter



change in NbSe₂ are reported to be 0.56 K/GPa and 16.2×10⁻³/GPa [17]. Combining these values, we expect T_c change as much as 0.017 K at $B_{\Phi} = 8$ T, which is negligibly small.

Figure 3. (a) Magnetic hysteresis loops for NbSe₂ single crystal irradiated by 800 MeV Xe with matching field $B_{\Phi} = 0.5$ T. The inset shows the data for a pristine sample. (b) Magnetic field dependence of J_c at T = 2 K for pristine and irradiated NbSe₂ samples. (c) J_c as functions of B_{Φ} at 2 K and at B = 0.1 T, 0.2 T, and 0.5 T. (d) The normalized pinning force density (F_p/F_{pmax}) calculated using $F_p(B) = J_c \times B$, at 2 K for NbSe₂ single crystals irradiated by 800 MeV Xe.

Magnetic hysteresis loops at various temperatures after 800 MeV Xe irradiation with $B_{\Phi} = 0.5$ T are shown in figure. 3(a). The inset shows a magnetic hysteresis loop for a pristine NbSe₂ sample at T = 2K. Compared with the pristine NbSe₂ sample, the hysteresis loop becomes much broader after 800 MeV Xe irradiation, indicating that effective pinning centers are introduced. A preliminary TEM observation on NbSe₂ irradiated by 800 MeV Xe shows very faint columnar defects with a lower density expected from B_{Φ} . We speculate that the threshold for the creation of columnar defects in NbSe₂ is very close to the energy deposition rate of 800 MeV Xe in NbSe₂ of 2.27 keV/ Å. It is known that columnar defects are not introduced in iron-based superconductors by 800 MeV Xe irradiation [9] since its energy deposition rate is below the threshold of the creation of columnar defects.

The evolution of the J_c as a function of B_{Φ} at T = 2 K is summarized in figure. 3(b). After the irradiation by 800 MeV Xe, the J_c in NbSe₂ is strongly enhanced. Even at $B_{\Phi} = 0.5$ T, the J_c increases significantly compared with the pristine sample. At small B_{Φ} , the J_c increases steadily with B_{Φ} up to 4 T. Above $B_{\Phi} = 4$ T, the J_c starts to decline. It should be noticed that, for all the irradiated samples, a

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weak peak around zero field gradually develops (vertical arrows in figure. 3(b)). One possible origin of this peak is the self-field effect [9]. That is, when the external field is smaller than the self-field, vortices in the superconductor will be strongly curved, when the applied field exceeds the self-field, all these curved vortices will be straightened, and hence the J_c is enhanced at around the self-field. The B_{Φ} dependence of J_{c} at several external fields are shown in figure. 3 (c), which shows a more clearly the behavior of J_c as a function of B_{Φ} . Figure. 3 (d) compares the normalized pinning force density (F_p/F_{pmax}) calculated using $F_p(B) = J_c \times B$ as a function of the reduced field $h = B/B_{irr}$ for different B_{Φ} at T = 2 K, where B_{irr} is the irreversibility field defined by the criterion of $J_c = 50$ A/cm². As described in Dew-Hughes's model [18], the pinning can be expressed as $F_p(h) \propto B_{c2}^n(T) f(h)$, where f(h)changes depending on the character of the pinning mechanism. According to the Dew-Hughes's model, the *h* value that gives a maximum of F_p changes for different pinning mechanisms. For example, in the case of surface pinning $f(h) = h^{1/2} (1-h)^2$ and $h_{\text{max}} \sim 0.2$, while in the case of point pinning $f(h) = h(1-h)^2$ and $h_{\text{max}} \sim 0.33$. In the case of NbSe₂ after irradiation by 800 MeV Xe, the peak shifts from $h_{\text{max}} \sim 0.23$ ($B_{\Phi} = 0.5$ T) to 0.36 ($B_{\Phi} = 8$ T). In cuprate superconductors irradiated by heavy-ions, various scaling functions are reported [19, 20]. For example, in YBa₂Cu₃O₇ thin film irradiated by 0.86 GeV Pb, a peak at h_{max} ~0.33 is reported, while in a similar film irradiated by 5 GeV Xe shows a peak appears at $h_{\text{max}} \sim 0.5$. These differences depend on the form of introduced defects and the preexisting defects in the sample. In the present case of NbSe₂, we can ignore the effect of pre-existing defects. However, the morphology of defects is not clear yet. A detailed study combined with highresolution STEM observations will shed light into the pinning mechanism in the irradiated NbSe₂ samples.

4. Summary

Strong pinning centers have been successfully introduced in NbSe₂ single crystals by 800 MeV Xe irradiation. Defects introduced by 800 MeV Xe irradiation up to $B_{\Phi} = 8$ T have no obvious effects on T_c , while *c*-axis lattice parameter increases steadily with the density of defects. J_c is found to be enhanced dozens of times by the 800 MeV Xe irradiation compared with the pristine sample. This indicates the NbSe₂ single crystal itself is a quite clean system, and also the defects introduced by 800 MeV Xe irradiation have strong positive effects on the enhancement of J_c .

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