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Simulation of Light C 4+ Ion Irradiation and Its

Enhancement to the Critical Current Density in

BaFe 1.9 Ni 0.1 As 2 Single Crystals

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ABSTRACT

In this work, we analyse the inﬂuence of C4+ irradiation with ion ﬂounce of 3×10 12 up to 2.3×10 15 ion·cm−2 on

signiﬁcant enhancement of the critical current density,Jc, in BaFe1.9Ni0.1As2 single crystals. Jc was increased

from 0.61×105 up to 0.94×105 A/cm2 at T=10 K and H=0.5 T. BaFe1.9Ni0.1As2 single crystals with and without the C4+-irradiation were characterized by magneto-transport and magnetic measurements up to 13 T over a wide range of temperatures below and above the superconducting critical temperature, Tc. It is found that the C4+-irradiation causes little change in Tc, but it can greatly enhance the in-ﬁeld critical current density by a factor of up to 1.5. Higher dose of C4+ions, causes further Jc enhancement at T=10 K. furthermore, ﬂux jumping completely disappeared at T=2 K after second C4+-irradiation. Our Monte Carlo simulation results show that all the C4+ions end up in a well deﬁned layer, causing extended defects and vacancies at the layer, but few defects elsewhere on the irradiation paths. Furthermore, the normal state resistivity is enhanced by the light C4+-irradiation, while the upper critical ﬁeld, Hc2 , the irreversibility ﬁeld, Hirr, and Tc were affected very little.

KEYWORDS:

critical ﬁeld (H c2 ) and nearly isotropic superconducting 5 Since the discovery of superconductivity in LaFeAsO, 1

properties as a result of small coherent length (). 4 One there has been an enormous deal of research on the super-

conductivity in the iron based superconductors in order to of the main requirements for practical application of the

understanding their properties and to investigate the poten-

122-FeAs superconductors is to carry high supercurrent

tial for applications.AFe 2 As 2 (A=Ba, Ca, Sr) supercon-

in high magnetic ﬁelds. One way to improve supercur-

ducting compounds have attracted great interests for the

rent is by creating defects deliberately in the crystal lat-

study of superconducting properties as they have simple

tice which can act as a strong pinning centre for pin-

structures and large single crystals can be grown. 2 The

ning vortices. There have been several studies that have

un-doped BaFe 2 As 2 (122-FeAs) shows a structural phase

aimed to enhance the effective defects and as a result

increase pinning in iron pnictide superconductors. For

netic transition from paramagnetic to spin density wave

example, the effect of neutron irradiation for SmFeAsO 1−x

state. 3 Superconductivity can be realized if the low temper-

sample shows thatJ cincrease by a factor of 3 atT=

ature phase transition is suppressed via chemical doping

5 K, 6 however it depress theT c. It is well known that

either on the Fe, Ba or As sites or by hydrostatic pressure.

columnar defects created by heavy ion irradiation are the

122-FeAs Family exhibits intermediate superconducting

most effective pinning sites to pin two-dimensional (2D)

transition between that of conventional lowT csupercon-

pancake vortices in highly anisotropic high temperature

ductors and highT ccuprate, 4 very highJ c, high upper

cuprate superconductors 78 pnictide superconductors have

revealed much smaller anisotropy (=1–8 atT≈T c), 59

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especially in doped BaFe 2 As 2 (122) superconductors with

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≈1–3. Also very strong intrinsic pinning strength has

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been observed in K doped 122 single crystals with rigidAccepted: 2 February 2014 2-February-2014

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vortices, mainly due to small anisotropy. 10 As a result, the

point defects induced by neutron irradiation are effective

for pinning vortices and enhancing the critical current den-

sity,J c, by a factor of 1.5–3. 11 Heavy ion irradiation using

ions such as Au, 12 Pb, 1314 and Ta 15 increasesJ cby a fac-

tor of 3–10 due to the formation of columnar defects. 1215

Both neutron and heavy ion irradiation are expensive pro-

cedures compared to light ion irradiation for large-scale

applications. Here, we report the ﬁrst efforts to create

defects by light-ion, C 4+, irradiation into optimally Ni

doped BaFe 19 Ni 01 As 2 single crystal. The inﬂuence of C 4+

irradiation on the physical properties of BaFe 19 Ni 01 As 2

single crystal has been investigated. Our results show that

light carbon ion irradiation is an effective approach that

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Fig. 1.Carbon ion distribution in the sample after C 4+-irradiation.

can signiﬁcantly enhance in-ﬁeldJ cwith little change in

T cwith ion ﬂounce of 3×10 12 up to 23×10 15 ion·cm−2 .

We also found that ﬂux jumping disappeared completely

after high dose irradiation. Furthermore, the Monte Carlo

simulation indicates that the C irradiation only cause dis-

tortions to the 122 lattice at a well-deﬁned layer, causing

little change to the lattice along its irradiation paths.

Single crystals with the nominal composition

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BaFe 19 Ni 01 As 2 were prepared by a self-ﬂux method. 16

The as-grown single crystals were cleaved and shaped into

thin plates for measurements. Irradiation with 35.59 MeV

C 4+was carried out perpendicular to the broad surface

of the sample, using a square shaped beam 7×7 mm 2 in

cross-section, for a total irradiation time of 3 min with

ion ﬂux of 3×10 12 ions·cm−2 . Second irradiation was

performed using ion doze of 23×10 15 ion·cm−2 . For the

sake of consistency, all the irradiation and measurements

were carried out on the same piece of single crystal sam-

ple. The sample was placed on a conductive sample holder

with conductive C-tape, in order to prevent charging and

excessive heating during irradiation. The beam current

was measured before and after irradiation with a Faraday

cup, and the average beam current was approximately

10 nA. The GEANT 4 package was used for the Monte

Carlo calculations to estimate the distribution of carbon

ions and the redistribution of other ions caused by carbon

ion collisions. Magnetization was measured using a mag-

netic properties measurement system (MPMS, Quantum

Design). The critical current density was calculated using

the Bean model. The transport properties were measured

over a wide range of temperature and magnetic ﬁelds up

to 13 T with applied current of 5 mA using a physical

properties measurement system (PPMS, Quantum Design).

Figure 1 shows the distribution of carbon ions in the

BaFe 19 Ni 01 As 2 single crystal using the Monte Carlo cal-

culation. The results show that almost all the C ions end

up in a well deﬁned layer, at a depth of around 24m.

This layer looks quite homogenous for 500 carbon ions

ﬁred along the red arrow. As the beam of carbon ions is

uniformly distributed across the sample surface, we expect

a fairly homogenous distribution of carbon in this layer.

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The binding energy of BaFe 19 Ni 01 As 2 is about 3 eV/atom,

so most of the damage is done by primary carbon ions

through primary knock-on collisions and none by the Ba,

Fe, Ni, and As recoils, because their energy is below 3 eV,

as shown in Figure 2(a).

The energy carried by the C 4+ions into the irradiated

layer is distributed to the BaFe 19 Ni 01 As 2 crystal lattice,

and as a result, the atoms in that layer will recoil or be

moved out of their lattice sites. Some of these atoms will

fall back into a thermodynamic equilibrium position (self-

annealing), but a number of them will remain in interstitial

positions, destroying locally the BaFe 19 Ni 01 As 2 lattice. To

see which of the lattice atoms are more disrupted by the

carbon ions, the calculated distributions of the individual

atoms (Ba, Fe, Ni, As) which are knocked out of their

lattice sites are shown in Figure 2(b).

Figure 2(b) shows that most of the BaFe 19 Ni 01 As 2 lat-

tice disruption is contained in and around the C 4+-irradiated

layer, at a depth of around 24m, with little disruption

between the entry surface and the damaged layer. Also,

the most disrupted (recoiled) are Fe and As, due to their

having the highest concentrations and lower masses. The

total number of vacancies produced by C-ions and the Ba,

Fe, Ni and As recoils is around 2,300 vacancies/ion in

the damaged layer. According to this calculation, the C-

irradiation and the resulting C-irradiated layer constitute

a three-dimensional (3D) defect layer with a thickness of

1.5m at a depth of about 24m under the irradiated

surface. The distribution of damage in the cross-section of

this 3D layer has a Gaussian proﬁle. This damage matrix

is likely to form a network (connected regions) in the dam-

aged layer. Therefore, the defect/vacancy region coexists

with the superconducting region which was not destroyed

during C-irradiation. This type of defect distribution, which

is very similar to extended defects, is distinct from the

columnar defects caused by heavy ion irradiation. 12

The temperature dependence of the resistivity at zero

magnetic ﬁeld for the sample before and after C 4+-

irradiation is shown in Figure 3. The resistivity decreases

with decreasing temperature from 300 to 20 K for both

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Fig. 2.(a) The energy of the carbon ions is distributed to the atoms/ions in their paths through collisions. (b) The calculated distribution of the

individual Ba, Fe, Ni, and As atoms which are knocked out of their lattice sites.

an extended Bean model: 20m/ a1−a/3b (a < b),

wheremis the width of the magnetization loop, anda

from 143×10−5·cm to 31×10−5·cm after car- andbare the length and width of the sample perpendicu-

bon irradiation at 200 K, which is related to enhancement

of impurity scattering after C 4+-irradiation. The reduction

ofT cafter ion irradiation is a common feature observed

in many high-T cand pnictide superconductors, 611 since

it can be affected by different effects such as inter-band

scattering, 17 a reduction in anisotropy, 18 etc. However, the

C 4+irradiation only causes small changes in theT cand

transition width in our sample. TheT cwas 18.3 K with

a small transition width (T c) of 0.9 K for the sample

without irradiation. It decreased very little to 17.8 K with

almost the sameT c(0.8 K) after C 4+-irradiation at zero

ﬁeld (Fig. 3). The residual resistivity ratio,RRR=n

(300 K)/n(20 K), wherenis the normal state resistivity,

decreased from 1.97 to 1.88, indicating enhanced scatter-

ing centres after C 4+-irradiation.

Enhancement of vortex pinning by the light C 4+ion irra-

diation can be clearly seen from the magnetization mea-

surements. Figure 4(a) shows the magnetization curve at

2 K for the un-irradiated and irradiated sample. The mag-

netization in the irradiated sample is obviously enhanced.

J cwas calculated from magnetic hysteresis data using

Fig. 3.Temperature dependence of resistivity for zero magnetic ﬁeld.

The inset shows an enlargement of the transition region.

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lar to the applied magnetic ﬁeld, respectively. Figure 4(b)

shows the calculatedJ cfor pristine and C 4+-irradiated sin-

gle crystal using different ﬂounce as a function of ﬁeld

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withHc. The irradiated sample shows a clearly enhanced

J c, which is both ﬁeld and temperature dependent. AtT=

10 K, theJ cis enhanced forH <4 T using 3×10 12

ions·cm−2 ﬂounce. ForT=2 and 5 K, theJ cenhancement

persists in both low and high ﬁelds.

Higher ﬂounce of 23×10 15 ion·cm−2 irradiation led

to two important consequences. Firstly, the ﬂux jumping

at 2 K disappeared completely. Secondly, theJ cenhanced

by a factor up to 1.5 for zero ﬁeld. The zero ﬁeldJ c

increased up to the high value ofJ c=72×10 5 using

the high irradiation ﬂounce. The peak effect, which has

been commonly observed in the Fe-based superconductors,

was observed for both irradiated and un-irradiated sam-

ples (Fig. 4(b)). The peak position shifts to lower magnetic

ﬁeld using low ﬂounce ion irradiation and move to higher

magnetic ﬁeld for ion ﬂounce of 23×10 15 , as indicated

by the arrows in Figure 4(b).J cis as high as 16×10 5

A/cm 2 at 5 K atH=05 T before C 4+-irradiation. The

J cincreases to 23×10 5 A/cm 2 after using 3×10 12 ions·

cm−2 ﬂounce. It has been reported that for BaFe 18 Co 02 As 2

crystals irradiated by neutrons with a ﬂounce of 4×10 17

cm−2 , 11 theJ cincreased from 3×10 5 to 7×10 5 A/cm 2

atH=05 T. These results are comparable with those for

our C 4+-irradiated sample using much lower ion doses of

C 4+(10 12 /cm 2 ). Therefore, light C 4+-irradiation could be a

very effective and less expensive approach for enhancing

theJ cﬁeld performance in the Fe-based superconductors

for practical applications.

Another feature of BaFe 19 Ni 01 As 2 single crystal is that

the pristine and carbon irradiated single crystals show the

ﬂux jump effect, which is more pronounced in the irradi-

ated sample using 3×10 12 ions·cm−2 ﬂounce (Fig. 4(a))

as shown in the magnetization hysteresis loops at very

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Fig. 4.(a) Field dependence of magnetization at 2 K for un-irradiated and after ﬁrst and second irradiation; (b) the magnetic ﬁeld dependence of

critical current density at different ion ﬂounce and temperatures.

theH c2cwas only very slightly enhanced by the C 4+

is smaller than that observed in Ba 072 K 028 Fe 2 As 2 single

crystal, 1019 with the ﬂux lines fully penetrating into the

whole sample.

In order to further look into the effect of the C 4+-ion

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irradiation on other pinning related parameters such as the

upper critical ﬁeld,H c2 , the irreversibility ﬁeld,Hirr , and

the pinning potential, we have carried outR–Tmeasure-

ments in ﬁelds up to 13 T withHcorHab. Figure 5

shows theR–Tcurves for the BaFe 19 Ni 01 As 2 single crys-

tal before and after C 4+-irradiation withHab. TheT c

onset slowly shifts to lower temperatures with increasing

magnetic ﬁeld, which is related to the nearly isotropic

superconductivity in the 122 family at low temperatures. 5

H c2 is estimated as the ﬁeld at which the resistivity

becomes 90% of the normal state resistivity; whileHirr

is deﬁned by 10% of the normal state resistivity. The

H c2 in theabplane and along thecdirection is plot-

ted as a function of temperature in Figure 6. The esti-

mated slopes are−6.65 and−5.28 T/K forH c2 andHirr

before carbon irradiation, and they decline to−6.52 and

−4.64 T/K after C-irradiation inHab, respectively. The

slopes ofH c2 andHirr were 2.82 and 2.49 T/K forHc

before irradiation, and they change slightly to 2.9 and 2.03

T/K after irradiation, respectively. It should be noted that

Fig. 5.Temperature dependence of resistivity for different magnetic

ﬁelds with ﬁeld parallel to theab-plane before (right) and after (left)

carbon irradiation.

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irradiation. However, the other parameters were obviously

reduced. This is related to the reduction of the electron

mean free path due to increasing impurity scattering after

C 4+-irradiation.

Thermally activated ﬂux ﬂow (TAFF) is responsi-

ble for the broadening of the resistivity transition and

can be expressed by the following equation:T H =

nexpU0T H /k B T , wherenis the normal state resis-

tivity,k Bis Boltzmann constant, andU0 is the activation

energy. The best ﬁt to the experimental data yields a value

of the pinning potential (U0/k B) of 4100 K atH <1 T

for both irradiated and un-irradiated samples. TheU0/k B

values are shown in Figure 7. For comparison, we also

includeU0/k Bvalues for Ba 072 K 028 Fe 2 As 10 single crys-

2

tals. It can be seen that theU0 /kBfor BaFe 19 Ni 01 As 2 sin-

gle crystal is lower than the reported value of 9100 K

for Ba 072 K 028 Fe 2 As 2 single crystal forHab. 10 For both

BaFe 19 Ni 01 As 2 single crystals, the activation energy drops

very slowly with increasing applied magnetic ﬁeld forH <

1 T. It can be scaled asH−002 , and then decreases slowly

asH−09 forH >1 T forHab, This is in great contrast to

the nearly ﬁeld independentU0 in Ba 072 K 028 Fe 2 As 2 single

Fig. 6.Temperature dependence of the upper critical ﬁeld before and

after carbon irradiation for BaFe 2−xNixAs 2 single crystal.

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Fig. 7.Magnetic ﬁeld dependence of pinning potential for

BaFe 2−xNixAs 2 single crystal before and after carbon irradiation. Data

for Ba 072 K 028 Fe 2 As 2 single crystal were taken from Ref. [7].

crystals, 10 indicating different pinning mechanisms in the

Ni and K doped 122 single crystals in high ﬁelds.

It should be pointed out thatU0 is reduced for both

HcandHabin high ﬁelds after C 4+-irradiation. This

means that the pinning strength in the ion irradiated sam-

ple, which only reﬂects the pinning energy for ﬁelds close

toHirr and temperatures close toT c, is weaker compared to

the sample without irradiation. The observation of reduced

U0 at high ﬁeld can well account for the fact that the

C 4+ion irradiation does little to changeT c,H c2 , orHirr ,

however, it can enhance the in-ﬁeldJ csigniﬁcantly for

H < Hirr . Further investigation on theJ cenhancement is

underway using high C 4+ion doses and different energies

that can increase both defect density and create extended

defects at various irradiation depths in the 122 supercon-

ductors.

In conclusion, we investigated the effects of C 4+-

irradiation in BaFe 19 Ni 01 As 2 single crystal. Monte Carlo

calculation shows that the C 4+ions end up in a well-

deﬁned layer at a certain depth, causing extended defects

and vacancies within the layer, but few defects elsewhere

on their paths. It is found that the C 4+-irradiation causes

little change inT c, but it can greatly enhance in-ﬁeld crit-

ical current density by a factor of up to 1.5. Our results

suggest that light C 4+ion irradiation is an effective and

cheaper method for the enhancement ofJ cin pnictide

superconductors compared to the heavy ion irradiation and

neutron irradiation.

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