# Magnetostriction in the mixed state of superconducting 2*H*-NbSe<sub>2</sub> single crystals

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Magnetostriction measurements on 2H-NbSe<sub>2</sub> single crystals in the temperature range 1.5–8 K in a magnetic field up to 14 T are reported. Peak and oscillations in the measured field dependences of magnetostriction were observed near  $H_{c2}$ . The reversible and irreversible components are separated and analyzed in the region of peak. The scaling parameters are defined, the contribution of the elastic constants dependence on magnetic field is demonstrated. The oscillatory component is discussed regarding Landau quantization of electronic spectrum.

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#### Introduction

The recent magnetostriction measurements performed in the mixed state of superconductors, the so-called Shubnikov phase has proved to be the alternative tool for studying interplay between the Abrikosov flux line lattice (FLL) and crystal lattice [1-4]. Though unambiguous manifestation of pinning phenomena in the irreversible magnetostriction was obtained some questions are still open. Among them is a discrepancy of model calculations and experimental data for high- $T_c$  superconductors as well as the different orders of magnetostriction magnitude in high- and low- $T_c$  superconductors characterized by pinning forces of the same order, and the role of anisotropy. In this paper we report measurements on an anisotropic low-temperature superconductor 2H-NbSe<sub>2</sub>, known for its purity and excellent parameters, that make it nearly an ideal system to study the issues mentioned above [5]. The expectation is that in such a system one has a better chance of understanding the complex phenomena involved. In particular, we focus on the old and famous but poorly understood «peak effect» (i.e., a peak in the critical current slightly below  $H_{c2}$  that occurs in most classes of type II superconductors [6,7] including cuprates [8]). The 2H-NbSe<sub>2</sub> modification of niobium diselenide seems to be the best object for investigation of these problems, since its parameters of anisotropy are close to those of high- $T_c$  superconductors (see below), its crystal structure and vortex matter are well studied, and the large single crystals of excellent quality are accessible. In this paper we use a different approaches to this problem based on the irreversible magnetostriction measurements. The basic idea behind a magnetostriction (MS) experiment on a type II superconductor is that a change in field will cause extra forces to act on the fluxoids. These extra forces transmit to the lattice through the pinning centers. An increase in field creates new fluxoids at the samples surface. They will push the already present fluxoids deeper towards the center of the sample. If the time scale of the change of field is big enough for the situation of the fluxoids to be treated as quasi-static, the density of fluxoids (DOF) will be such that the extra compressive forces among fluxoids due to the field increase will be balanced by pinning forces. The DOF will have a decreasing slope going towards the center. The pinning centers will transmit these compressive forces to the sample. For the decreasing fields, a reverse process can be observed, resulting in a DOF with an increasing slope and an extra expansive forces on the sample. Because flux motion is irreversible the magnetostriction curves display a hysteresis and thus reveal the features of an interaction between the flux lines and the crystal lattice.

## System, sample preparation and experimental details

X-ray studies of the obtained NbSe<sub>2</sub> single crystals confirmed the 2*H* polytype. Magnetization studies of this crystal showed a sharp superconducting transition at  $T_c = 7.2$  K and  $\partial H_{c2} / \partial T = 8.1$  kG/K for the field parallel to the *c* axis. The transition width was found to be less than 20 mK at all fields. As in the magnetization studies, transport measurements found  $\partial H_{c2} / \partial T = 7.9$  kG/K. For  $H \parallel c$ , the transport critical current was somewhat higher than that inferred from magnetization hysteresis, ranging up to 40 A/cm<sup>2</sup> at low fields. The critical current showed a very pronounced peak effect near  $0.9H_{c2}$  as has been reported previously [5]. Below the following denotations will be used:

 $\lambda_{\uparrow} = \Delta L_{\uparrow} / L$  is the length change in increased field;

 $\lambda_{\downarrow} = \Delta L_{\downarrow} / L$  is the length change in deacreased field;

 $\lambda_p = \Delta L_{\uparrow} / L - \Delta L_{\downarrow} / L$  is the irreversible (pinning induced) magnetostriction;

 $\lambda_{\rm rev} = (\Delta L_{\uparrow} / L + \Delta L_{\downarrow} / L)/2$  is the reversible part of magnetostriction;

 $\lambda(c, a), \quad \lambda_p(c, a), \quad \lambda_{rev}(c, a) \text{ stand for the } \Delta L/L \parallel c \text{ and } H \perp c;$ 

 $\lambda(c, c), \lambda_p(c, c)$  stand for the  $\Delta L/L \parallel c$  and  $H \parallel c$ ;  $\lambda(a, a), \lambda_p(a, a)$  stand for the  $\Delta L/L \perp c$  and  $H \perp c$ ;

 $\lambda_{pm}$  is the peak value of irreversible magnetostriction;

*H* is the external magnetic field;

 $H_{c2}$  is the upper critical field;

 $H^*$  is the field of irreversibility dissapearance (peak goes to zero);

 $h = H/H^*$  is the reduced magnetic field;

 $F_n$  is the pinning force;

 $F_s^{P}$  is the shear force of FLL (flux line lattice);

 $j_c$  is the critical current density;

C is the elastic modulus.



Fig. 1. Measured magnetic field dependences of magnetostriction  $\lambda(c, a)$  in 2*H*-NbSe<sub>2</sub> single crystal in a field normal to the hexagonal axis ( $H \perp c$ ). The magnetostrictive length change was measured along the *c*-axis. The arrows ( $\rightarrow$ ) indicate the dependences obtained during magnetisation and the arrows ( $\leftarrow$ ) correspond to reverse process (*a*, *b*). Longitudinal magnetostriction  $\lambda(a, a)$  versus applied magnetic field normal to the hexagonal axis ( $H \perp c$ ) (*c*, *d*). Longitudinal magnetostriction  $\lambda(c, c)$ versus magnetic field applied along the hexagonal axis ( $H \parallel c$ ) at T = 4.2 K (*e*).

The length changes were measured using a parallel-plate capacitance method. The direction in which the length changes were measured was parallel or perpendicular to the field direction. The length change measurements of  $\lambda(c, a)$ ,  $\lambda(c, c)$ , and  $\lambda(a, a)$  were performed. The field sweeps were never faster than 0.18 T/min.

#### **Experimental results**

Here we present the results for the  $\lambda(c, a)$ (Fig. 1,*a*,*b*),  $\lambda(a, a)$  (Fig. 1,*c*,*d*), and  $\lambda(c, c)$ (Fig. 1,*e*) measurements. Orientation of field in *ab*-plane of 2*H*-NbSe<sub>2</sub> requires field up to 14 T in order to reach the  $H_{c2}$  below 2 K. The  $\Delta L$  measurements in *ab*-plane require capacitance cell of special design and high sensitivity.

Figure 1,*a*,*b* presents the measured field dependences of magnetostriction  $\lambda(c, a)$  in 2*H*-NbSe<sub>2</sub> single crystal in a magnetic field normal to the hexagonal axis  $(H \perp c)$ . The magnetostrictive length changes were measured along *c*-axis. In Fig. 1,*c*,*d* some typical MS curves measured in *ab*-plane with longitudinal orientation of the applied magnetic field for different fixed temperatures are presented. A «dip» for increasing and a «peak» for decreasing fields is observed. In Fig. 1,*e* longitudinal magnetostriction  $\lambda(c, c)$  versus magnetic field applied along the hexagonal axis is presented at the temperature 4.2 K. The irreversible component of magnetostriction  $\lambda_p$  was separated and is shown in Fig. 2,*a*,*b*. The measurements have given the ratio  $\lambda_p(a, a)/\lambda_p(c, a) \geq 10^2$ .

#### Discussion

#### Irreversible magnetostriction

Previous works [1–4] describe how the vortex flux pinning by the lattice is clearly shown from magnetostriction measurements at the superconducting regime, as we believe to have observed in the present 2H-NbSe<sub>2</sub> also. First of all and most important is the already mentioned strong hysteresis shown by the  $\lambda(c, a)$  and  $\lambda(a, a)$  modes, similar to the magnetization hysteresis, which is assigned to pinning effects. Second, as it is well known, the critical current density  $j_c$  is a rapidly decaying function of both field and temperature. From Fig. 2,*a*,*b* we can notice that low temperature irreversible magnetostriction isotherms show a field dependence with a sharp maximum at a certain field, typical for the systems with weak pinning on point defects [9] which increases with decreasing temperature, reminiscent of the mentioned  $j_c$  beha-



*Fig.* 2. Irreversible part of magnetostriction measurements at different temperatures, *T*, K: 1.5 (1); 2.5 (2); 4.2 K (3):  $\lambda_p(c, a)$  (a);  $\lambda_p(a, a)$  (b).

vior. The relation between pinning force (flux gradient) and lattice deformation in the specified direction x may be obtained from the equation of elastic balance [3]:

0

$$\Delta L \cong 1/C \int_{0}^{L/2} F_p \, dx \,. \tag{1}$$

In Eq. (1) L is the dimension of our platelet samples along the selected direction ( $L \sim 1.32$  mm along *c*-axis and  $L \sim 5$  mm in *a*-direction). The field and temperature dependences of  $F_p$  can be well explained by using Kramer's [7] theory of pinning forces for type II superconductors, but introducing a slight modification in order to apply such a theory to superconductors with a pronounced peak effect below  $H_{c2}$ . In terms of reduced magnetic fields  $h = H/H^*$  for  $h << h_{pm}$  ( $h_{pm}$  corresponds to the peak value of irreversible magnetostriction  $\lambda_{pm}$ ) flux motion primarily occurs by depinning of the weakly pinned vortices present in the sample, and for  $h >> h_{pm}$  flux motion occurs by synchronous shear of the vortex lattice around



*Fig.* 3. Irreversible magnetostriction  $\lambda_p$  normalized to its peak value versus a reduced magnetic field  $h = H/H^*$ :  $\lambda_p(c, a)/\lambda_{pm}(c, a)$  (a),  $\lambda_p(a, a)/\lambda_{pm}(a, a)$  (b).

strong pinning centers. To these two regimes the pin-breaking pinning force,  $F_p$ , and the vortex lattice shearing pinning force,  $F_s$ , correspond [7,9]. In this way the models [7,9] yield scaling law  $F_p(H) \sim f(h)$ , and predict a  $(H^*(T))^{2.5}$  dependence for  $F_p$ . Thus one should expect for the flux-pinning magnetostriction the scaling behavior followed by  $F_p$  (see (1)), if the field dependence of elastic constants is not taken into account. This approach perfectly describes the results obtained on the high- $T_c$  superconductors [1].

In Fig. 3,*a*,*b* we plot the ratio  $\lambda_p(c, a)/\lambda_{pm}(c, a)$ and  $\lambda_p(a, a)/\lambda_{pm}(a, a)$  versus  $H/H^*$ , the data points collapsing to universal curves as also expected from [7]. The  $H^*$  values corresponding to temperatures below 4.2 K were adjusted to get the best collapse of the respective  $\lambda_p/\lambda_{pm}$  isotherms in the universal curve. At the same time we were unable to explain the observed scaling just by using the  $H^{*2.5}$  law predicted by [7], i.e. in our case the logarithmic plot  $\lambda_{pm}$  versus  $H^*$  gives the slope of about 6 instead of the expected 2.5.

Therefore, following [2] we were overlooking the fact that from a macroscopic thermodynamic argument, a modified Ginzburg–Landau description [10], it can be shown that the elastic constant in the superconducting state at  $H_{c2}$  is proportional to the normal state elastic constants times  $(H_{c2})^{-2}$ and elastic modulus in (1)  $C(T, H) = C_0(h)(H_{c2})^{-2}$ with  $C_0(h)$  being a function depending on h (con-



*Fig.* 4. Scaling plot for irreversible magnetostriction:  $\lambda_n(c, a)$  (*a*),  $\lambda_n(a, a)$  (*b*).

taining also the normal state elastic constants). The above considered dependence of the macroscopic pinning force density on magnetic field indicates that  $\lambda$  should obey the scaling law  $\lambda = H_{c2}^{4.5}g(h)$  in which g(h) is a function only dependent on h. Taking into account the above mentioned arguments for substitution of  $\lambda$  by its irreversible part  $\lambda_p$ , and  $H_{c2}$  by  $H^*$ , where irreversibility disappears, we plotted  $\lambda_p / (H^*)^{4.5}$  as a function of  $h = H/H^*$  for low temperatures in comparison with the plot  $\lambda_p / (H^*)^{2.5}$  suggested before (see Fig. 4). Indeed the scaling law is closer to the experimental curve but the best fit is obtained for  $\lambda_p(c, a)/(H^*)^{6.5}$  and  $\lambda_n(a, a)/(H^*)^6$ .

In conclusion, the above results point to a vortex lattice pinning origin for the irreversible strains observed at low enough temperatures for the studied 2H-NbSe<sub>2</sub> samples. The important conclusion is that the magnetostriction measurements represent an alternative tool, on top of the usual one from magnetization and critical current measurements. In the present measurements the scaling behavior of magnetostriction in a range of the peak observed near  $H_{c2}$  satisfies scaling low similar to that for pinning force. The models of pinning explain the results on magnetostriction in the mixed state of 2H-NbSe<sub>2</sub> if the dependence of elastic modulus on a magnetic field is taken into account. The specifics of the 2H-NbSe<sub>2</sub> layered structure should be adhered to a large power magnitude.

#### Reversible magnetostriction. Quantum oscillations

We have reported on the peculiarities of the irreversible magnetostriction component in the mixed state of the superconducting single crystals 2H-NbSe<sub>2</sub>, which was determined as the difference between magnetostriction values during magnetisation and those during demagnetisation of the sample in external magnetic field. Now we consider the reversible part of magnetostriction which is determined as the sum between magnetostriction values during magnetisation and those during demagnetisation of the sample in an external magnetic field. The reversible part  $\lambda_{rev}(c, a)$  of  $\lambda(c, a)$  includes two components. The first one is the monotonic function of a magnetic field and the other component is oscillatory function of magnetic field. The oscillating component unambiguously results from the Landau quantisation of electron spectrum, being analog of the de Haas-van-Alphen (dHvA) effect, which was observed earlier in the mixed state of 2H-NbSe<sub>2</sub> [11].

Magnetic field dependences of magnetostriction and its reversible part are presented for the same



*Fig.* 5. Field dependences of the reversible part of  $\lambda(c, a)$  magnetostriction measurements. The arrows show peak position for  $\lambda$ .

temperatures in Figs. 1 and 5, which clearly demonstrate an existence of oscillatory component in field dependence of  $\lambda_{rev}(c, a)$ . The oscillatory part of  $\lambda(c, c)$  is possibly not observed because of large



*Fig.* 6. Oscillatory magnetostriction component of  $\lambda_{rev}(c, a)$  as a function of inverse magnetic field. The arrows indicate the peak position for  $\lambda$ .

frequency period according to Fermi surface calculations [11]. For the case of  $\lambda(a, a)$  the experimental error is too high to observe the oscillations.

Magnetostriction dependences on an inverse magnetic field (Fig. 6) contain mostly a single harmonic similarly to the dHvA effect [11]. As in the dHvA effect the amplitudes of oscillations below  $H_{c2}$  and above it slightly differ, which is attributed to the additional electron scattering on the Abrikosov flux lines in the mixed state of superconducting 2*H*-NbSe<sub>2</sub> [12].

The formula of Lifshits—Kosevich [13] for the oscillating thermodynamic potential, which was obtained in assumption of the arbitrary dispersion law allows one to consider both dHvA oscillations and those of magnetostriction. Chandrasekhar [14,15] has shown that amplitude of the oscillations of magnetostriction and of absolute magnetisation values coincide within a factor  $\partial \ln (S_m)/\partial \sigma_{\alpha}$ , where  $S_m$  is the extreme cross section area of the Fermi surface normal to the direction of a magnetic field,  $\sigma_{\alpha}$  is the stress along this direction. The parameter  $\partial \ln (S_m)/\partial \sigma_{\alpha}$  is relevant to the theory of ultrasonic attenuation due to the electrons, and is closely analogous to deformation potential in semiconductors.

Using our data on magnetostriction together with the data on the absolute magnetisation values would allow us to find the magnitude of  $\partial \ln (S_m) / \partial \sigma_{\alpha}$ . However such data for 2*H*-NbSe<sub>2</sub> have not been described in literature by now. We plan to present these results in the nearest publication.

#### Monotonic component of reversible magnetostriction and thermal expansion

Reversible magnetostriction consists of two parts. The first one is determined by volume increase of the normal component in the mixed state  $V_N(H)$  and volume decrease of the superconducting one  $V_S(H)$  during increase of applied field. The second part is magnetostriction of the normal component itself.

Therefore at fixed temperature we obtain

$$\lambda_{\text{rev}} = \delta_T \frac{V_N(H)}{V_N(H + V_S(H))} + \lambda_N(H) . \qquad (2)$$

Here  $\delta_T$  is the difference of the sample sizes in the superconducting and normal states at fixed temperature  $T < T_c$ . Figure 7 presents a temperature dependence of this variable, which was obtained as a difference between thermal expansion in superconducting state ( $T < T_c$ ) and that in the normal



*Fig.* 7. Temperature dependence of the exceeding thermal expansion along c axis in the normal state over that in the superconducting state.

state at the same temperature T. The latter was obtained by extrapolation of the experimental data to the range of low temperatures as it is shown in Fig. 8.

Magnetic field dependence of the longitudinal magnetostriction  $\lambda(c, c)$  in the mixed state of 2H-NbSe<sub>2</sub> is shown in Fig. 1,*e* for field applied along *c*-axis at 4.2 K. This is the most understandable situation as magnetic susceptibility and magnetostriction in normal state  $(T > T_c)$  are negligibly small at this orientation according to anisotropy of Fermi surface. Regarding the first part in equation (2) one can easily estimate  $\lambda(c, c)$  if  $\delta_T$  at 4.2 K is known. In a result one obtains, that  $\lambda(c, c)$  approaches the magnitude  $\delta_T = 6.5 \cdot 10^{-7}$  in the fields  $H > H_{c2} = 2.3$  T for these orientation of magnetic field and T = 4.2 K. This magnitude is close to the measured one.



*Fig.* 8. Experimental curve of thermal expansion along hexagonal *c*-axis (solid curve) and extrapolation of thermal expansion into the normal state in a range  $T < T_c$ .

#### Conclusions

1. In a temperature range below critical temperature magnetostriction of 2H-NbSe<sub>2</sub> reveals both reversible and irreversible parts.

2. In magnetic fields corresponding to a peak on the field dependences of critical current a dip in increased field and a peak during demagnetization are observed.

3. In reduced coordinates  $\lambda_p / \lambda_{pm} = f(H/H^*)$  the irreversible part of magnetostriction is described by universal dependence for different temperatures  $T < T_c$ . Here  $\lambda_{pm}$  is the peak value of irreversible magnetostriction in the field  $H^*$  above which irreversible magnetostriction disappears.

4. In the coordinates  $\lambda_p / (H^*)^n = f(H/H^*)$  the measurements of irreversible magnetostriction correspond satisfactorily to universal curve at n = 6, which differs from the expected value n = 4.5.

5. At a field orientation perpendicular to the *c*-axis the field dependence of the reversible magnetostriction along *c*-axis reveals an oscillatory component. Amplitude of this component is strongly periodic with respect to inverse magnetic field 1/H. This part of magnetostriction is explained by Landau quantization of electronic spectrum and is similar to de Haas—van Alphen's effect (dHvA).

6. The linear thermal expansion demonstrates slight peculiarity at  $T_c$ . At  $T < T_c$  the monotonic part of reversible magnetostriction saturates at fields above  $H_{c2}$  and is much higher than magnetostriction in the same fileds though at  $T > T_c$ . This part of reversible magnetostriction is explained by increase of the volume of normal component in the mixed state during an increase of the external magnetic field.

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