

On the transverse magnetization of the anisotropic superconductor $2H\text{-NbSe}_2$

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Torque measurements were performed on a high-quality single crystal of the uniaxial superconductor $2H\text{-NbSe}_2$ in a tilted magnetic field 0–200 kG, in the temperature range 1.5–4.2 K. The transverse component of the absolute magnetization was derived in a magnetic field directed at an angle of 77° to the axis of anisotropy, and its field dependence was analyzed in a reversible domain of the mixed state. The penetration depth and anisotropy characteristics were obtained for the sample under study.

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1. Introduction

Measurements of the transverse magnetization of magnetically anisotropic media have proved to be an effective tool for the study of spatially inhomogeneous magnetic states. As a result of such investigations V. V. Eremenko and co-authors have discovered intermediate [1] and mixed [2] states in antiferromagnets, like those existing in type-I and type-II superconductors, respectively. Torque measurement is a technique for registration of the transverse magnetization of a crystal in a uniform magnetic field, which has been used successfully in studies of the magnetic anisotropy [3] (cited from [4]) and the Fermi surface [4] for a long time. The torque value τ in Gaussian units is determined by the simple equation

$$\tau = -M_{\perp} H V, \quad (1)$$

where H is the magnetic field, V is the sample volume, M_{\perp} is the transverse (with respect to the applied magnetic field) component of the absolute magnetization, $M_{\perp} = M_x \cos \theta - M_z \sin \theta$, z is the direction along c , x is the direction along the a or b axis in the ab plane, i.e., normal to the c axis, and θ

is the angle between the c axis and the external magnetic field direction (insert in Fig. 1).

Observation of a sample's torque suggests a non-zero component of the magnetization off from the magnetic field direction and, consequently, an orientational dependence of the sample's free energy in a magnetic field. Torque measurements on conventional superconductors [5–7] were made in connection with an investigation of magnetic flux pinning by crystal lattice inhomogeneities. Since the discovery of high-temperature superconductivity (HTSC) and the observation of the orientation of fine particles of these anisotropic compounds in a magnetic field [8–10], the possibility of intrinsic torque in anisotropic superconductors in the region of equilibrium magnetization has been assumed [11]. Experimental evidence for the existence of transverse magnetization and intrinsic torque of anisotropic HTSC was obtained for the first time in [12]. Later on, intrinsic torque measurements were performed on single crystals of HTSCs [13–15] and organic superconductors [14]. In those works the thermodynamic properties of the equilibrium Abrikosov vortex lattice were examined in the framework of London electrodynamics on the basis of an-

gle dependence of the torque (Kogan model [11]) and Ginzburg–Landau phenomenology (Clem model [16,17]). In the present work the field dependences of the equilibrium torque are analyzed for the first time for the uniaxial superconductor $2H\text{-NbSe}_2$.

2. Experimental part

The measurements aimed at observation of transverse magnetization in equilibrium region of superconducting mixed state placed the following specific requirements on the samples and experimental techniques employed. 1) The sample should be an anisotropic superconductor. Our sample under study meets this requirement, as its anisotropy parameters are close to those of the yttrium HTSC, which was the first object used for reversible torque observation (the ratios of effective masses along the c axis and in the superconducting plane are 10 and 25, respectively, which means that both of them are moderately anisotropic superconductors). Moreover, the superconducting compound $2H\text{-NbSe}_2$ is characterized by uniaxial anisotropy, and, consequently, one does not need to assume negligibility of the orthorhombicity in the crystallographic planes responsible for superconductivity. 2) Pinning effects should be minimal. Actually, the high-quality single crystals of $2H\text{-NbSe}_2$ under study are characterized by a ratio of critical currents attributed to pinning and to depairing of $j_c / j_0 \approx 10^{-6}$, which describes them as the cleanest type-II superconductors. Their field dependences of the magnetization have two reversible regions, viz., weak fields $H \ll H_{c2}$ and near the upper critical field H_{c2} ([18] and the references therein). 3) As the magnitude of equilibrium magnetization is not large compared to the applied field (see, e.g., [19]), the torque should be amplified by the sample's volume according to (1). This was realized in our case due to the opportunity of obtaining large crystals of $2H\text{-NbSe}_2$. The volume of the crystal under study was $19.7 \cdot 10^{-9} \text{ m}^3$, which was estimated to an accuracy of better than 1% by weight measurements (127 mg) and using the density value 6.44 g/cm^3 , calculated from the atomic distribution in the unit cell of $2H\text{-NbSe}_2$, determined from the x-ray measurements. 4) According to the published data on the magnetization of $2H\text{-NbSe}_2$, the torque signal near the upper critical field H_{c2} should be of the order of $10^{-3} \text{ dyn}\cdot\text{cm}$ ($10^{-10} \text{ N}\cdot\text{m}$), if the transverse magnetization were equal to full magnetization. The available torque measurement technique [20] gave an accuracy of $10^{-5} \text{ dyn}\cdot\text{cm}$. It was achieved in the following way. The torque on the

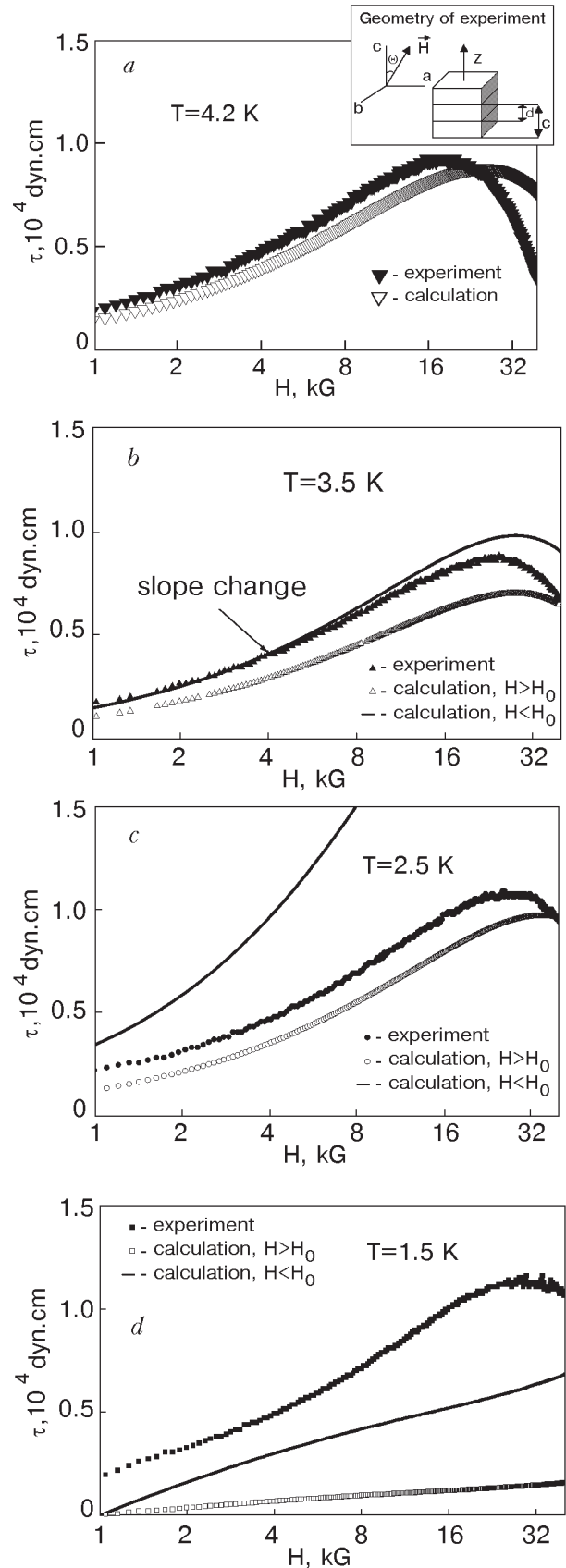


Fig. 1. The measured and calculated dependences $\tau(H)$ for $T = 4.2$ (a); 3.5 (b); 2.5 (c); 1.5 (d) K, plotted in semilogarithmic scale. (Insert: c is the crystal lattice spacing and d is the interlayer distance.)

sample was transmitted to the registering unit by a Cu-Be spring, the rigidity of which was chosen taking into account weight of the sample. The signal was registered by the capacitance method of Griessen [21] using a bridge scheme. This method allowed us to measure the capacitance with an accuracy of 10^{-4} pF. The large-scale magnet M7 at the High Magnetic Field Laboratory in Grenoble provided a highly uniform field in the sample region. Calibration of the signal was performed using a miniature coil (diameter 4.8 mm, length 1.4 mm, number of turns 20) mounted in the region of the sample in a field of 12 T, to an accuracy of 3%. The out-of-plane angle of the external field direction was 23° , which was estimated with an accuracy of 1° , satisfactory for measurements of the magnetic-field dependence of the torque.

The experimental geometry is presented in Fig. 1, *a*. The measurements presented in this figure (a typical field dependence of the torque, measured at a temperature of 4.2 K) clearly show the occurrence of transverse magnetization of the $2H\text{-NbSe}_2$ single crystal in the field range $H_{c1} \ll H \ll H_{c2}$, where H_{c1} is the lower critical field. A thermodynamic data analysis follows below.

3. Discussion of experimental results

The measured field dependences of the torque are presented in Fig. 1 in a comparison with those calculated using the equation [11]

$$\tau(H) = \frac{\Phi_0 H V}{64\pi^2 \lambda_{\alpha\beta}^2} \frac{\gamma^2 - 1}{\gamma^{2/3}} \frac{\sin^2 \theta}{\varepsilon(\theta)} \ln \left[\frac{\gamma \eta H_{c2} (||c)}{H \varepsilon(\theta)} \right], \quad (2)$$

where Φ_0 is the magnetic flux quantum, and ε and γ are the anisotropy parameters,

$$\varepsilon(\theta) = (\sin^2 \theta + \gamma^2 \cos^2 \theta)^{1/2}, \quad (3)$$

and $\eta \sim 1$.

Relation (2) was derived on the basis of London electrodynamics. It was already proved by experiments on HTSCs with moderate (Y-Ba-Cu-O) and strong (Ba-Bi-Ca-Cu-O and Tl-superconductors) anisotropy [12,13]. The compound under present study is a classic London superconductor [22], as the magnetic field penetration depth λ in it sufficiently exceeds its coherence length ξ (for the component of M normal to the ab plane, which is responsible for superconductivity, $\lambda_{ab} \approx 1000$ Å and the coherence length $\xi_c \approx 10$ Å although it is nevertheless larger than the interlayer distance). A satisfactory agreement between the measured and calculated dependences is observed in a limited low-field range. The field of apparent deviation of the mea-

sured curve from the calculated one (the field of the maximum in the field dependence of the torque) is a function of temperature. This may suggest that the field H_{int} is related to the influence of intervortex interactions on the magnetization. It can be calculated taking into account that this interaction becomes considerable when the vortex lattice spacing equals to the penetration depth:

$$a_0 = \sqrt{\frac{\Phi_0}{H_{\text{int}}}} = \lambda. \quad (4)$$

3.1. Transverse component of equilibrium magnetization

Though the occurrence of the transverse component of the equilibrium magnetization of uniaxial superconductors was predicted before the discovery of HTSC, and the estimates were made for $2H\text{-NbSe}_2$ [23] itself, until now the related measurements had been performed on HTSCs only. The satisfactory agreement between the measurements and calculations in this work may be used for estimation of the thermodynamic properties of the compound under study. Initially the transverse magnetization will be determined on the basis of London electrodynamics and the restrictions for its application will be established.

In Gaussian units [19]

$$G_i = F_i(T, B_i) - \frac{B_i H}{4\pi}, \quad (5)$$

where F_i and G_i are the free energy and Gibbs potential for the specific volume of the i th phase (below, the indices i and j correspond respectively to the mixed state or Shubnikov phase and to the phase with $B \equiv H$, where the major part of the sample remains in the normal state).

The relation between field and induction in the mixed state follows from the condition of minimum of G at fixed H and T :

$$\frac{\partial}{\partial B_i} F_i(T, B_i) = \frac{H}{4\pi}. \quad (6)$$

The thermodynamic Gibbs potential for the system of interacting vortices is determined by the following equation:

$$G = n_L \mathcal{F} + \sum_{i,j} U_{ij} - \frac{BH}{4\pi}, \quad (7)$$

where \mathcal{F} is the free energy of an individual vortex, n_L is the number of vortices per unit volume, U_{ij} is the potential of repulsive interaction between vortices, and

$$B = n_L \Phi_0 . \quad (8)$$

Therefore, the first term in (7) is the sum of the individual vortex energies.

The second term in (7) describes the energy of repulsive interaction between the vortices. It will be examined below for different field ranges, following [11,23–29].

The third term in (7) considers influence of an applied field H . It tends to favor large values of the induction B . This means that the field H stands for an external pressure which tends to increase the vortex density.

The following analysis of the measurements will use a quantitative estimate of the vortex interaction in the range $H_{c1} \ll H \ll H_{c2}$ or $\xi \ll a_0 \ll \lambda$ (ξ is the superconducting coherence length, $a_0 = \sqrt{\Phi_0/B}$ is the vortex lattice spacing, and λ is the penetration depth), or $1/\lambda^2 \ll n_L \ll 1/\xi^2$. In this range of fields and vortex lattice parameters of a *uniform* superconductor in the London limit ($\lambda \gg \xi$) the free energy is equal to [19]

$$F = \frac{B^2}{8\pi} + \frac{B}{4\pi} H_{c1} \frac{\ln(\beta a_0/\xi)}{\ln(\lambda/\xi)} , \quad (9)$$

where β is a parameter that depends on the vortex lattice symmetry. For a triangular lattice $\beta = 0.381$. Considering this relation, the equation for the magnetization was obtained [26]:

$$-4\pi M = (\Phi_0/8\pi\lambda^2) \ln(H_{c2}\beta/H) . \quad (10)$$

In this equation β is determined from experimental data. Actually the field range considered [26] is narrower, as it is restricted below by the field $H' > H_{c1}$ above which the irreversibility effects may be neglected, i.e., relation (10) is fulfilled in a field range of

$$H' < H \ll H_{c2} . \quad (11)$$

The London equations may be generalized to the case of a uniaxial anisotropic superconductor through substitution of the isotropic effective mass by an anisotropic mass tensor [23]. Then Eq. (9) transforms to the free-energy equation for a vortex lattice at an angle θ to the symmetry axis c [27]:

$$8\pi F = B^2 + (\Phi_0/4\pi\lambda^2) \times \\ \times (m_1 B_x^2 + m_3 B_z^2)^{1/2} \ln(H_{c2}\beta/B) , \quad (12)$$

where $m_1 = m_2$ are the components of the mass tensor along the in-plane crystallographic axes ab , m_3 is that along the c axis. Minimization of (12) with

respect to B gives [26] the magnetization components:

$$-M_z = M_0 \frac{m_3 \cos \theta}{\sqrt{m(\theta)}} , \quad -M_x = M_0 \frac{m_1 \sin \theta}{\sqrt{m(\theta)}} , \quad (13)$$

where

$$M_0 = \frac{\Phi_0}{32\pi^2\lambda^2} \ln \frac{H_{c2}\beta}{H} , \quad (14)$$

$$m(\theta) = m_1 \sin^2 \theta + m_3 \cos^2 \theta .$$

An expression for the magnetization components measured in a direction perpendicular to the applied field was also derived in [26]:

$$M_{\perp} = M_x \cos \theta - M_z \sin \theta = \\ = M_0 \frac{m_3 - m_1}{\sqrt{m(\theta)}} \sin \theta \cos \theta . \quad (15)$$

3.2. Transverse magnetization of the crystal under study

The measured dependences $M_{\perp}(H)$ are presented in Fig. 2. The slope $M_{\perp}(\ln H)$ allows one to find the magnetic-field penetration depth λ , using Eqs. (13) and (14). Applying the following relation, which follows from London theory,

$$\lambda_{ab} = m^* c^2 / 4\pi n_s e^2 , \quad (16)$$

one obtains $m^* \approx 7m_e$. Here m_e is the mass of a free electron, and n_s is the density of superconducting electrons. For the sample with $\lambda = 1200$ Å the value $m^* = 13.5m_e$ [30] was obtained. The cyclotron mass obtained from analysis of the high-field oscillation amplitude with the lowest frequency (which corresponds to the in-plane field orientation), following

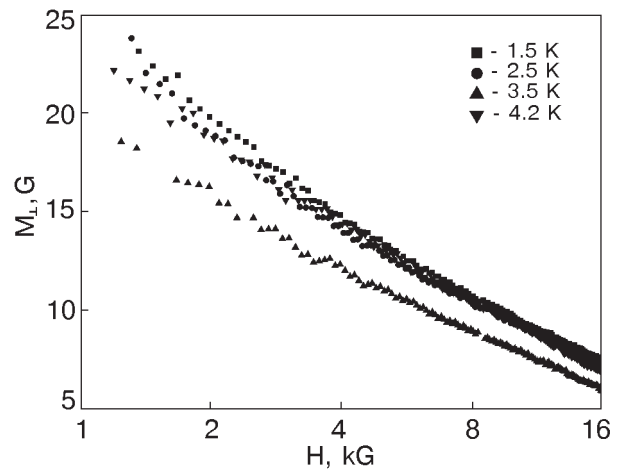


Fig. 2. Field dependences of the transverse component of the magnetization $M_{\perp}(H)$ in a semilog plot, calculated from experimental data presented in Fig. 1.

[31], appeared to be lower by an order of magnitude: $m^* \sim 0.6m_e$.

Thus the cyclotron mass m^* at a field orientation $H \perp c$ is close to the lower component of the mass tensor m_1 , while the mass derived from the penetration depth of the field normal to the plane of the sample, i.e., $H \parallel c$, is close to the large component of mass tensor m_3 .

3.3. Restrictions of London electrodynamics application

The London model is in reality responsible only for the logarithmic prefactor $-M_0$ and for the linear dependence of M on $\ln H$ in (13). The factor at H_{c2} under the logarithm appears initially as a fitting parameter. In such a form the $M(\ln H)$ dependence allows one to obtain reasonable values of H_{c2} from the field dependences of equilibrium magnetization in a temperature range $T/T_c \approx 0.4 - 1$ [32]. The same is true for comparison of our torque measurements with equation (2): a satisfactory agreement is observed down to $T \approx 2.5$ K (for the material under study $T_c = 7.2$ K) (see Fig. 1).

The limitation of London electrodynamics is connected with the fact that it does not consider the contribution of the energy of the vortex cores (suppression of the order parameter) to the total energy of the Abrikosov vortex lattice. The situation can be improved at the moment in two ways: 1) using a procedure proposed in [16,17], which is based on the phenomenological Ginzburg–Landau approach and is strictly valid near the upper critical field and in the vicinity of T_c , which is not the case here; 2) taking into account the nonlocal correction after the method described in [32]. That is important for estimation of H_{c2} from magnetization measurements. In the lower limit of the intermediate fields near T_c the calculations of the torque function $\tau(H)$ by techniques based on the Ginzburg–Landau and London models practically coincide (see Fig. 3). That corresponds our data on $\tau(H)$ at $T = 4.2$ and 3.5 K (Figs. 1,a,b). It should be noted that $T \approx 3$ K is the quantum limit temperature for our sample ($T_{ql} = T_c^2/E_F$, where E_F is the Fermi energy), below which the nonlocality radius $\rho(T)$ must be an important parameter in all of the microscopic calculations [33].

The temperature dependence of the field H_0 at which the nonlocal effects «switch on» (Fig. 4), according to [32] can have minimum; that is consistent with our data. In [32] the dependence $H_0(T) = \Phi_0/2\pi^2\sqrt{3}\rho^2$ was derived for materials with a cylindrical Fermi surface, which is the case for $2H\text{-NbSe}_2$ (see, e.g., [31]). Unfortunately in

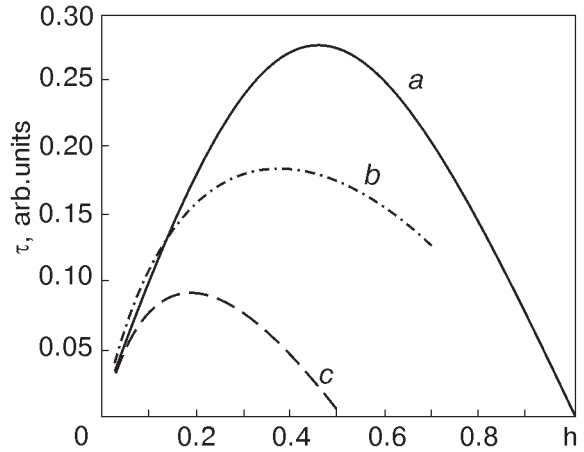


Fig. 3. Calculated torque as a function of $h = H/H_{c2}$: from Ginzburg–Landau model [16] (a); from London model [11] with $\eta=1$ (b); from London model [11] with $\eta=0.5$ (c).

our case an exact estimation of $H_0(T)$ is not possible, because of the lack of reliable data on the electron mean free path in our sample. According to early data on this material [30], the most optimistic estimates give its value as 250 \AA . At the same time, estimation of this characteristic from the oscillatory component of the reversible magnetization M for a sample with all the measured parameters being similar to ours gives $\sim 1000 \text{ \AA}$ [31]. If these data are true, we are working with an even cleaner ($l \gg \xi$) single crystal than suggested. Then the possible contribution from nonlocal effects should increase.

Taking into account that the magnetostriction follows the same laws as the magnetization and using equations [26] for the magnetization components along the principal crystallographic axes together with our data on magnetostriction along a and c (Fig. 5) [34] one can obtain independently

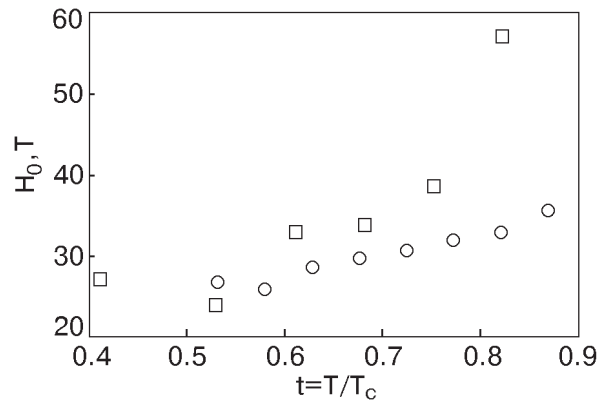


Fig. 4. Temperature dependence of nonlocality parameter $H_0(T/T_c)$ of anisotropic superconductors: Bi-2212 (\square), Tl-2212 (\circ) [32].

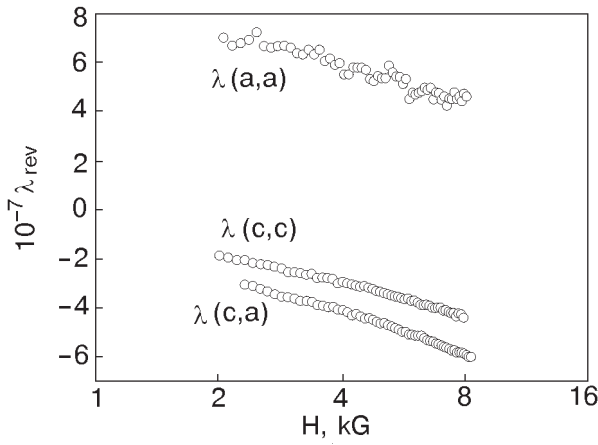


Fig. 5. Field dependence of the reversible magnetostriction $\lambda_{\text{rev}}(H)$ in a semilog plot: $\lambda(a,a)$, $\lambda(c,a)$ – field applied along a axis; $\lambda(c,c)$ – field applied along hexagonal axis.

the ratio of the anisotropic mass tensor components:

$$\sqrt{\frac{m_3}{m_1}} = \frac{dM(0)/d \ln H}{dM(\pi/2)/d \ln H} = \frac{d\lambda_{\text{rev}}(0)/d \ln H}{c_{11}} \bigg/ \frac{d\lambda_{\text{rev}}(\pi/2)/d \ln H}{c_{33}} \approx 3, \quad (17)$$

where λ_{rev} is the reversible magnetostriction, c_{11} and c_{33} are the components of the elastic modulus. Thus m_3 is an order of magnitude higher than m_1 .

4. Summary

1. The torque measurements were performed on single crystal of $2H\text{-NbSe}_2$ superconductor with moderate anisotropy. In a limited range of intermediate fields the measurements are well described by Kogan's formula, derived on the basis of London electrodynamics. This torque is connected neither with pinning nor with a form factor, but results from the tendency of Abrikosov vortices (the normal to circular currents, screening vortex cores) in anisotropic superconductor to build up in a preferred crystal direction.

2. The transverse component of magnetization is isolated.

3. The penetration depth λ is calculated and compared with published data. The data obtained for λ are used for estimation of the effective electron masses, which are compared with the data on the cyclotron mass. The latter was determined from analysis of the amplitudes of magnetization oscillations with the lowest frequencies. The cyclotron mass m^* in a field $H \perp c$ is close to the small component m_1 of the mass tensor, and the mass determined from the penetration depth for out-of-plane

magnetic field $H \parallel c$ is close to the large component m_3 of the mass tensor. The magnitudes of the effective electron masses determined from the penetration depths for magnetization oscillations in a field oriented at angle 77° to the crystal axis c differ by an order of magnitude. That corresponds to the ratio of the components of the anisotropic mass tensor, derived from magnetostriction measurements.

4. Comparison of the field dependences of the magnetostriction and magnetization allowed us to estimate the anisotropy parameter as $\gamma = 3$, i.e., the ratio $m_3/m_1 \sim 9$ from magnetostriction measurements in fields directed along the crystal axes.

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