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Magnetization and magnetostriction oscillations in a superconducting $2H$-NbSe$_2$ single crystal

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The results of a comparative study of the oscillating magnetostriction and absolute magnetization components in the single crystal of the layered superconducting compound $2H$-NbSe$_2$ are presented. The measurements were made in the temperature range 1.5–8 K in a magnetic field of 0–20 T with in-plane orientation (normal to the hexagonal axis). The mechanical stress derivative of the extremal cross-sectional area of the Fermi surface is derived from a thermodynamic analysis. The values obtained are considerably higher, by an order of magnitude, than those for other metals, which explains the observed enhancement of the magnetostriction oscillations in comparison with the magnetization oscillations.

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1. Introduction

L. V. Shubnikov was the first one, who has observed the oscillations of resistivity [1]. Exactly this observation lead to discovery of the de Haas–van Alphen (dHvA) oscillations of the magnetic properties. The layered dichalcogenide $2H$-NbSe$_2$, with a transition to the charge-density-wave (CDW) state at temperature $T_{CDW} = 32$ K, is the first superconducting compound (the superconducting transition temperature $T_c = 7.2$ K) in which Landau oscillations of the electron spectrum have been observed [2]. It was the first observation of the de Haas–van Alphen and magnetothermal oscillations not only in the normal state but also in a mixed state, the so-called Shubnikov phase ($H < H_{c2}$). The measurements were made at temperatures much lower than T_c for different orientations of the external magnetic field **H** with respect to the crystallographic axes of the single crystal under study. In [2] and the subsequent investigations [3–7] of superconducting $2H$-NbSe$_2$ the measurements of the magnetization oscillations were made at a temperature far below 1 K in order to eliminate the electron scattering, as above the Dingle temperature ($T_D = 2.5$ K [5], $T_D = 1$ K [6] for $2H$-NbSe$_2$) it reduces drastically the amplitude of the oscillations, and their registration by existing techniques becomes impossible. Measurements in magnetic fields below the upper critical field H_{c2} have shown that in the mixed state the oscillations are suppressed by the scattering of normal (quantized) electrons on Abrikosov vortices, observed in other superconductors also (see the review [8] and the references therein). The measured angular dependences of the oscillation frequency $F(\theta)$, where θ is the angle between the hexagonal axis and field direction, at $H < H_{c2}$ revealed a pronounced dependence $F(\theta)$ in the angle range $0 < \theta < 20°$ and a weak dependence $F(\theta)$ at $\theta > 60°$ [6]. The band calculations [5] demonstrate that the measured angular dependence of oscillations is well described in terms of the «small flat hole Fermi surface around the Γ point». The above-mentioned investigation complemented the results of previous band calculations of the Fermi surface in $2H$-NbSe$_2$ [9–11]. Existing data about the shape and peculiarities of Fermi surface in $2H$-NbSe$_2$ suggest its high sensitivity to pressure and stress, which has been proved by the measured pressure dependences of the superconducting parameters of this compound ([12–14] and references therein). The observed magnetostriction oscillations in superconducting single crystals of $2H$-NbSe$_2$ [15] in comparison with dHvA oscillations would allow one to study the influence of mechanical stresses on the band structure of this material according to the thermodynamic approach developed in [16–18].

We should mention, that in [19] the deformation effect on dHvA oscillations is considered. The related change of oscillations frequency is estimated. The deformations of 10^{-2}–10^{-3} are taken into account. Here, we analyse the amplitudes ratio for magnetization and magnetostriction, the amplitude of the latter being of the order 10^{-5}. The analysis of frequencies will be presented elsewhere.

In this work a comparative analysis of the oscillatory magnetostriction (λ_{osc} and absolute magnetization $\mathbf{M} = \mathbf{m}/V$ ($\mathbf{m}$ is the magnetic moment of the sample and V is its volume) measurements is performed. The same single crystal of $2H$-NbSe$_2$ was studied by capacitive dilatometer and capacitive cantilever torquemeter [20] techniques.

2. Experimental techniques

The absolute magnetization measurements were performed on single crystals of the layered compound $2H$-NbSe$_2$ (the space group $P6/mmm$ or D^4_{6h}). The high anisotropy of crystal structure ($a = 3.45$ Å, $c = 12.54$ Å) is accompanied by anisotropy of electronic properties, in particular, of the superconducting parameters: $H_{c2}(T = 4.2\ \mathrm{K}) \approx 7$ T for $\theta = 90°$ and 2.3 T for $\theta = 0°$ [2,15,21,22] (there is some discrepancy in the published data). The mass of the sample was $m = (127 \pm 0.05)$ mg. The sharp superconducting transition was observed at temperature $T_c = 7.23$ K. The detailed information on the sample preparation technique and characterization is presented in [15]. In that paper the first observation of magnetostriction oscillations in the mixed state of any superconductor was presented. The measurement of magnetostriction $\lambda^{(c,a)}$ was performed on a single crystal of $2H$-NbSe$_2$ in the crystallographic direction (001), i.e., the c axis, in a magnetic field parallel to the a axis. The oscillatory component of $\lambda^{(c,a)}$ was defined as $\lambda^{(c,a)}_{\mathrm{osc}} = \lambda^{(c,a)}_{\mathrm{rev}} - \lambda^{(c,a)}_{\mathrm{mon}}$, where $\lambda^{(c,a)}_{\mathrm{rev}} = (\lambda_{\mathrm{up}} + \lambda_{\mathrm{down}})/2$ is the reversible part of $\lambda^{(c,a)}$; $\lambda^{(c,a)}_{\mathrm{up}}$ is $\lambda^{(c,a)}$ measured during magnetization of the sample; $\lambda^{(c,a)}_{\mathrm{down}}$ is $\lambda^{(c,a)}$ measured during demagnetization of the sample; $\lambda^{(c,a)}_{\mathrm{mon}}$ is the monotonic part of $\lambda^{(c,a)}_{\mathrm{rev}}$.

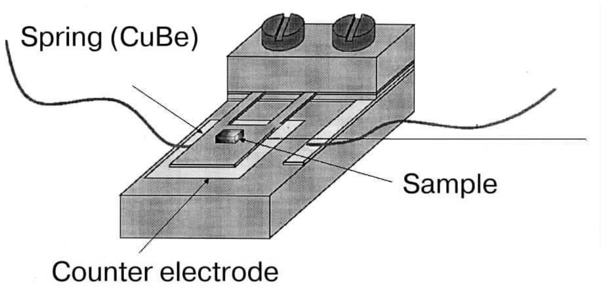

Fig. 1. Capacitive torquemeter for magnetization measurements. The calibration coil is positioned in the sample area (not shown here).

In the present work the absolute magnetization measurements were performed in a temperature range 1.5–4.2 K in a magnetic field up to 20 T using a capacitive cantilever torquemeter (see Fig. 1). The field sweep rate was 0.18 T/min. The torque magnetization measuring technique [22] is based on the fact that in a magnetic field $\mathbf{H}$ a magnetized sample experiences a torque $\mathbf{T}$ proportional to the component of the sample's magnetic moment perpendicular to $\mathbf{H}$:

$$\mathbf{T} = \frac{V}{\mu_0}\,\mathbf{M}\times\mathbf{H}\ , \tag{1}$$

where μ_0 is the vacuum magnetic permeability.

In a sample with an anisotropic Fermi surface the magnetization component $M_\perp$ perpendicular to the field direction is proportional to the parallel component $M_\parallel$ [23]:

$$M_\perp = -\,kM\ ,\ k = \frac{1}{\mathcal{F}}\frac{\partial \mathcal{F}}{\partial \theta} \tag{2}$$

where $\mathcal{F}$ is the free energy.

The torque $\mathbf{T}$ results in deflection of a Cu–Be spring joined with the sample. The change of the distance between the spring and adjacent metallic plate is measured by a capacitance technique. At $\theta = 90°$ the measured signal from torquemeter disappears, as k becomes equal to zero. For this reason the field dependence of $\mathbf{M}$ was measured at $\theta = (77 \pm 1)°$. The measurements revealed equal frequencies of the magnetization and magnetostriction oscillations, $F_M = F_{MS}$, which agrees with the weak angular dependence $F(\theta)$ at $60° < \theta < 90°$, measured before. In order to find the absolute values of $\mathbf{M}$ the calibrations of the measuring capacitance were performed in each experiment using a calibrating coil, which was positioned in the region of the sample (Fig. 1). The volume V of the sample was determined using the density value $\rho = 6.3 \cdot 10^3$ kG/m^3, calculated from the known atomic distribution in 2H-NbSe$_2$ [5]. The oscillatory component of M was defined in the same way as the oscillatory component of the magnetostriction: $M_{\text{osc}} = \frac{1}{2}(M_{\text{up}} + M_{\text{down}}) - \overline{M}$, where $M_{\text{rev}} = = \frac{1}{2}(M_{\text{up}} + M_{\text{down}})$ is the reversible component of M, M_{up} is the absolute magnetization of the sample measured in increasing magnetic field after zero field cooling (ZFC); M_{down} is the absolute magnetization of the sample measured in increasing magnetic field; $\overline{M}$ is the monotonic component of M. In the oscillatory regime it was determined by averaging M_{osc}.

Analysis of M_{osc} was performed after subtracting the monotonic component according to [6].

3. Experimental results and discussion

Figure 2 presents a typical magnetic field curve of M_{rev} for a 2H-NbSe$_2$ single crystal, in comparison with λ_{rev} measured on the same crystal. It is clearly seen that in a region of the peak effect [15] both **M** and λ depend nonlinearly on **H**. This means that a simple summation of the dependences measured in increased and decreased fields does not completely eliminate manifestations of the peak effect on the $M_{\text{rev}}(H)$ and $\lambda_{\text{rev}}(H)$ curves. The oscillatory behavior of the measured dependences starts near H_{c2}. Unfortunately, because of the available experimental conditions the magnetostriction measurements were restricted by an upper value of the applied field $H = 14$ T, and comparison with the magnetization measurements is possible only for this range of H. The magnetization oscillations are less pronounced than those of the magnetostriction. They appear at higher fields, and their amplitude A_M is lower than the amplitude A_{MS} of the magnetostriction oscillations. Figure 3 presents the inverse field dependences of M_{osc} and λ_{osc} for $T = 1.5$ K, which were obtained by subtracting their monotonic components from M_{rev} and λ_{rev}, respectively. The insets show the results of a fast Fourier transform analysis of the measurements, which was used for determination of F_M and F_{MS}. It is seen that these frequencies coincide. The slight phase shift may be explained by the different geometry of the experiment, explained in the previous Section. The derived values of A_M and A_{MS} were used for calculations of $\partial(\ln S_m)/\partial\sigma_\alpha$, where S_m is the extremal cross-sectional area of the Fermi surface, and σ_α is the mechanical stress arising along the **H** direction. For that we used the expression

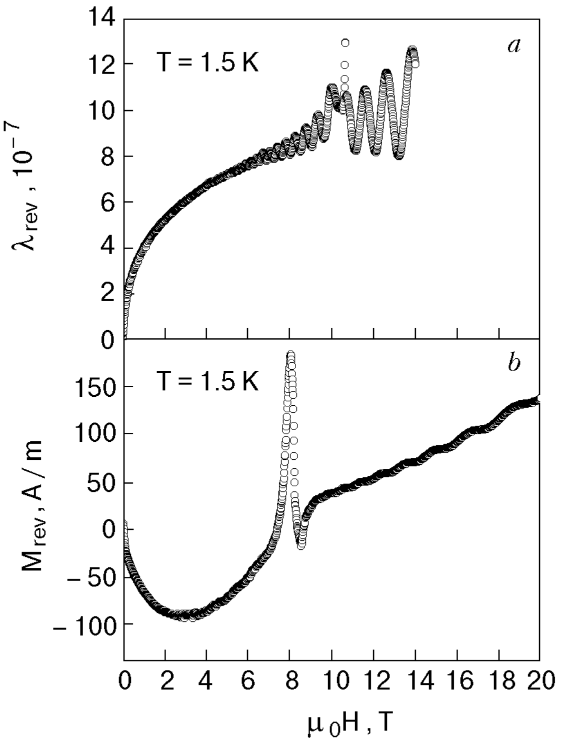

Fig. 2. The typical magnetic field dependences of λ_{rev} (*a*) and M_{rev} (*b*) for 2H-NbSe$_2$ single crystals.

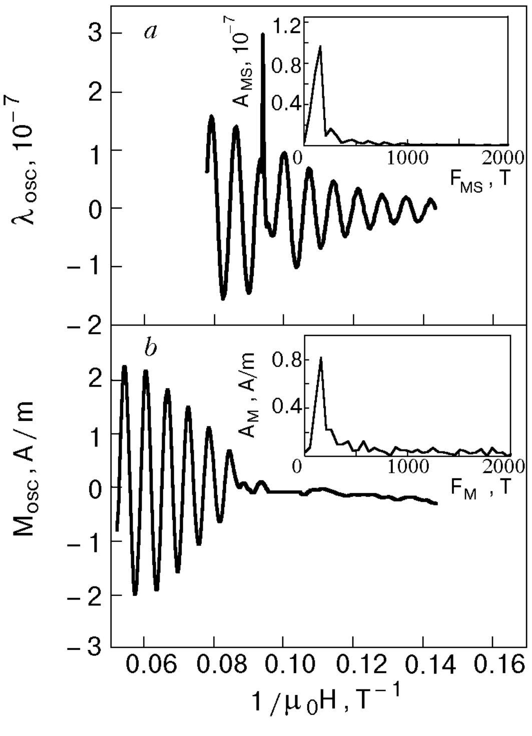

Fig. 3. The inverse field dependences of λ_{osc} (*a*) and M_{osc} (*b*) for $T = 1.5$ K. The inset shows the results of a fast Fourier transform analysis of the measurements, which was used for determination of F_M and F_{MS}.

$$\frac{\lambda}{\mathbf{M}\cdot\mathbf{H}} = \frac{\partial(\ln S_m)}{\partial\sigma_\alpha}\,, \tag{3}$$

which was obtained from a thermodynamic analysis of the oscillating part of the thermodynamic potential [16–18]. The results are presented in Table in comparison with the data from the review [18] for common metals.

Table

Comparison of $\partial(\ln S_m)/\partial\sigma_\alpha$ values for $2H$-NbSe$_2$ and other metals

Metal	$\partial(\ln S_m)/\partial\sigma_\alpha$ (in 10^{-11} m^2/N)
$2H$-NbSe$_2$	148.5 ± 11 [present work]
Cu	7.5 ± 2 [18]
Ag	24 ± 1 [18]
Au	8.3 ± 1 [18]

It is seen that the values of $\partial(\ln S_m)/\partial\sigma_\alpha$ obtained in the present work for $2H$-NbSe$_2$ are considerably, by an order of magnitude, higher than those obtained previously for the other metals. This is why the magnetostriction oscillations are enhanced in comparison with the magnetization oscillations.

Conclusions

A comparative analysis of the reversible absolute magnetization and magnetostriction as functions of the inverse magnetic field, measured on a superconducting $2H$-NbSe$_2$ single crystal, have shown that: 1) the oscillation periods M_{osc} and λ_{osc} coincide, which means that both effects are attributed to Landau quantum oscillations; 2) the magnetostriction oscillations are more pronounced than the magnetization oscillations: the former appear at lower fields, and their amplitude is higher (with respect to the monotonic component); 3) a thermodynamic analysis shows that the values of $\partial(\ln S_m)/\partial\sigma_\alpha$ for $2H$-NbSe$_2$ are much higher, by an order of magnitude, than the values of $\partial(\ln S_m)/\partial\sigma_\alpha$ for common metals. This is due to the peculiar geometry of Fermi surface in the highly anisotropic compound $2H$-NbSe$_2$ and explains the effect of magnetostriction «enhancement».

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