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THE HYSTERESIS OF ULTRASOUND ATTENUATION IN THE MIXED STATE OF NIOBIUM*

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In an impure crystal of Nb with a ⟨111⟩ orientation the hysteresis of complete and partial $S \rightleftarrows N$ transitions under the influence of the magnetic field ($H \| Q$) was studied by means of longitudinal and shear ultrasonic waves with the frequency $120 \div 550$ MHz. In the temperature range $1.8 \div 4.2$ K the characteristic fields including H_{c2} are linear functions of temperature. The lowering of temperature or the increasing of the field change rate enhances the occurrence of sudden changes of fluxoid population. The attenuation α characterizes unambigously the bulk condition of the mixed state.

ГИСТЕРЕЗИС УЛЬТРААКУСТИЧЕСКОГО ЗАТУХАНИЯ В СМЕШАННОМ СОСТОЯНИИ НИОБИЯ

Приведены результаты измерений гистерезиса переходов из нормального состояния в сверхпроводящее (и наоборот) в монокристалле ниобия с ориентацией ⟨111⟩ в магнитном поле с помощью продольных и поперечных ультразвуковых волн частоты 120–550 МГц. В области температур 1,8–4,2 К характеристические поля (включая H_{c2}) линейно зависят от температуры. Понижение температуры или повышение скорости изменения поля способствует появлению скачкообразных изменений числа флюксоидов. Затухание α однозначно характеризует смешанное состояние в объеме монокристалла ниобия.

I. INTRODUCTION

In superconductors of the second type the diamagnetic state persists up to the critical field H_{c1}. In higher fields the magnetic flux penetrates into the sample and the mixed state appears. In consequence of the negative interface energy $\sigma_{NS} = \xi - \lambda < 0$, (where ξ is the coherence length, and λ the penetration length) the interface area between the normal and the superconducting regions is enhanced, and a regular array of magnetic flux lines is formed. The mixed state exists up to the critical field H_{c2}, where the normal cores of fluxoids begin to overlap and the vortices disappear [1].

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The ultrasound makes it possible to study the mixed state of a superconductor as a bulk property even at zero induction. There is no unique theory of attenuation in the mixed state, as the energy dissipation has different reasons under different conditions, e.g. the purity (pure and dirty limit), the temperature ($T \ll T_c$, $T \leq T_c$), and the magnetic field ($H \geq H_{c1}$, $H \leq H_{c2}$) [2].

In real samples of finite dimensions the transition from the superconducting to the normal state is no more reversible due to the presence of surface barriers, which prevent the penetration or escape of flux lines [3] and of volume barriers restraining their movement [4]. The values of H_{c1} and H_{c2} are also changed by an unfavourable sample shape involving a high demagnetizing factor, and the edge effect (i.e. the deviation from the ideal elipsoidal shape). Gottlieb et al. [5] have studied the hysteresis of the $N \rightleftharpoons S$ transition in niobium of medium purity (RRR 700) at the frequency 510 MHz at about 7 K due to the magnetic field. The complex behaviour of this phenomenon led them to the opinion that the attenu- ation cannot be accounted for in terms of the microscopic theory [6].

In the present paper we shall describe experiments with a Nb single crystal of low purity (RRR 50) in the temperature range 1.8 ÷ 4.2 K at frequencies in the range 100 ÷ 550 MHz.

II. EXPERIMENTAL PART

The single crystal (denoted as A-1) used in this measurement was prepared in the Institute of Metal Physics of the Ukrainian Academy of Sciences by the electron zone melting method[1]). The original diameter 20 mm was lowered to 15 mm by electrolytical etching. The purity of the original material should have been quite high, but after mechanical grinding and polishing the RRR decreased to about 50, as revealed by the electrical resistance measurement in the longitudinal magnetic field on a wafer, spark cut out of the single crystal. According to [7], this ratio corresponds to the purity of about 99.94%. The axis of the single crystal deviated from the [111] direction by about 1.5°, and its length was 0.835 cm. The sample was not degassed or annealed before the measurement. The Matec ultrasonic equipment Model 6600 was used to excite nonmetallized quartz transdu- cers (supplied by Monocrystals Inc., Turnov) X — cut (13.6 MHz, diameter 5 and 6 mm) and Y-cut (13.6 MHz, 5×4 mm) for longitudinal and shear waves, respecti- vely. The automatic attenuation recorder Model 2470 A has proved to be useful while measuring the magnetic field dependence of ultrasound attenuation. The transducer bonded by Nonaq grease to the sample was placed in a vacuum

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[1]) I wis to thank Professor I. Ja. Dechtjar for the crystal.

chamber, in the compensated superconducting solenoid SM I. The external field strength was monitored by a LMK 85 Hall probe placed symmetrically to the sample with respect to the coil centre, about 40 mm apart from the sample, in an effort to remove the external field distortion caused by the sample itself; the demagnetizing factor of the sample was about $D \cong 0.8$ [8].

In our measurements the condition $Ql_e \ll 1$ was always fulfilled[2]).

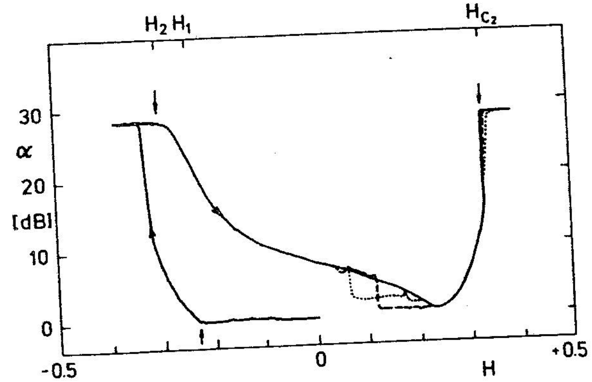

Fig. 1. Virgin transition $S \to N$ and hysteretic transition $N \to S \to N$ in the magnetic field $H \| Q \|$ [111] in the Nb single crystal A-1. Full line: $dH/dt = 0.58$ mT/s, dashed line 1.80 mT/s, and pointed line 7.75 mT/s. Frequency of longitudinal waves 540 MHz, temperature $T = 4.2$ K. (Measurement 20.12). The external field intensity is in $(10^7/4\pi)$ A/m $= 10^4$ Oe

III. RESULTS

For longitudinal waves of the frequency 540 MHz the results are shown in Fig. 1. During the virgin magnetization the attenuation $\alpha(H)$ hardly changes, until a certain magnetic field $H_1 = 0.24$ T is exceeded. Then a steep increase follows, and at about 0.33 T (which equals H_{c2}) the normal state is reached; this point is well pronounced. When we decrease the external field, the mixed state reappears at $H_2 = 0.30$ T, and the return branch does not reach zero at $H = 0$; the remanent attenuation α_R amounts to about $\frac{1}{3}(\alpha_N - \alpha_S)$. After reversing the external field direction, the curve continues to decrease until a minimum α_{min} is reached at $H_3 = 0.23$ T and then again an enhanced attenuation follows. For a low rate of $dH/dt = 0.58$ mT/s the curve is quasi-continuous; for higher rates cusps (jumps)

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[2]) The mean free path of the conduction electrons l_e varies as the residual resistance, i.e. $l_e = k$. RRR [μm]. Lee and Dobbs use $k = 2 \times 10^{-2}$ [9], whereas Forgan and Goubh [10] use $k = 4 \times 10^{-3}$. Taking $k = 1 \times 10^{-2}$, we get for our sample $l_e = 0.5$ μm. As long as $F < 1.5\Gamma$ Hz, the condition $Ql_e \ll 1$ is fulfilled ($Q = 2\pi/\Lambda$, where Λ is the wave length).

appear, connected either with the annihilation of fluxoids, having an opposite orientation, or with their escape from the sample. Also the shape of the curve near H_{c2} is rate dependent.

The attenuation in the superconducting state α_S serves as zero level for other measurements; it is predominantly influenced by the temperature. On the other hand, the attenuation in the normal state $\alpha_N(H_{c2})$ varies as frequency squared, T and H.

The temperature dependence of α for longitudinal waves has been measured at

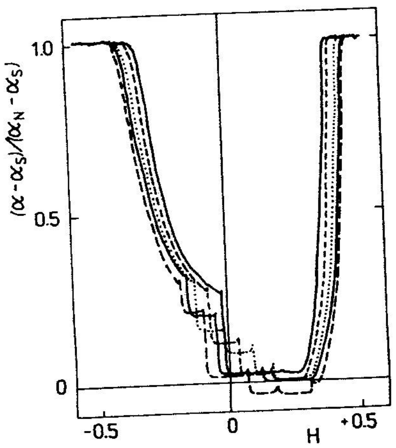

Fig. 2. Temperature dependence of the hysteretic $N \to S \to N$ transition in the magnetic field $H \| Q \|$ [111] of Nb single crystal A – 1. Frequency of longitudinal waves 480 MHz, temperatures ——— 3.44 K, - - - 2.98 K, ··· 2.48 K, ——— 2.10 K, –·– 1.79 K. dH/dt rate was 8.3 mT/sec. The ordinate is $(\alpha-\alpha_s)/(\alpha_N-\alpha_s)$ approximately. The attenuation α was measured in the single echo mode. At the temperature of 1.79 K an upward jump appeared. (Measurement 18.9). Field intensity in $(10^7/4\pi)$ A/m = 10^4 Oe

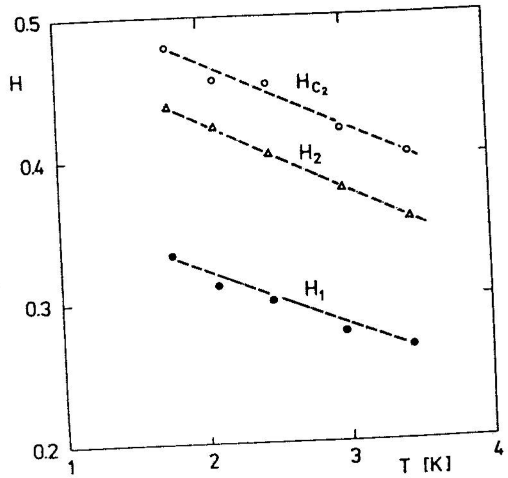

Fig. 3. The temperature dependence of H_{c2} (curve a), H_1 and H_2. These values are explained in Fig. 1. Other details are to be taken from Fig. 2, from which this one was redrawn. H is in units of $(10^7/4\pi)$ A/m = $= 10^4$ Oe

the frequency of 480 MHz. In Fig. 2 the relative attenuation was plotted with the same ordinate $\alpha_N(T) - \alpha_S(T)$ (not always succesfully) at a rate $dH/dt = 8.3$ mT/ /sec. At this rate the jumps of the attenuation appear near $H = 0$ at the temperature of 4.2 K, but with the lowering of the temperature the jumps appear at gradually higher fields before the zero field is reached. This phenomenon is due to the temperature dependence of the instabilities of the fluxoid array [10]. In Fig. 3 the temperature dependence of critical fields is shown, appearing quasi-linear in the narrow temperature interval studied.

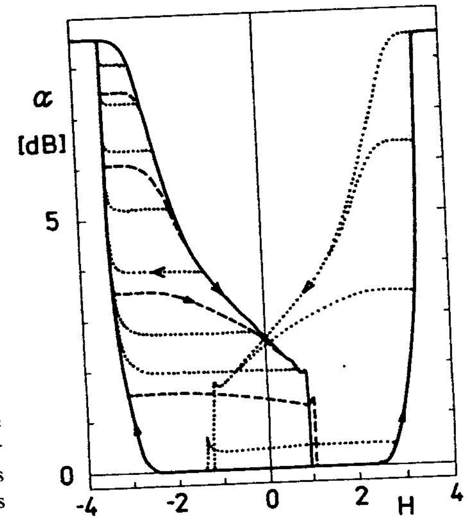

Fig. 4. Complete set of virgin, hysteretic and partial hysteretic curves of (α/H) in a Nb-single crystal A-1 at the temperature of 4.2 K. The frequency of shear waves was 121.5 MHz, for $\alpha(H)$ measurement the 1st and the 2nd echoes were used; dH/dt was 19 mT/s. (Meas. 23.7) H is in units of $(10^7/4\pi)$ A/m = 10^4 Oe

Besides the complete hysteresis also the partial hysteresis was studied, as shown in Fig. 4 for shear waves with the frequency 121.5 MHz, at the temperature of 4.2 K. All of them show a characteristic feature — after the sense of the field change is reversed, there is no change in the attenuation except in the near vicinity of the virgin curve (or near the hysteretic branch), which the partial hysteresis curve closely approaches.

The attenuation of shear waves shows a very steep increase of α when approaching H_{c2}, defining very distinctly this value. On the other hand, when the external field H is lowered, there is but a continuous change at $H < H_{c2}$. This is certainly caused by an inherent condition of the material, e.g. by the delayed penetration of fluxoids into the sample.

The attenuation was always amplitude independent; in the last experiment it has changed less than 10^{-2} dB/cm per 1 dB increase of amplitude.

IV. DISCUSSION

The unfavourable shape of our sample and the high demagnetizing factor have certainly worsened the homogeneity of the fluoxid distribution, the penetration of which is hampered by the deformed surface layer at the planparallel faces of the specimen. Fortunately, in fields near to H_{c2}, the homogeneity improves considerably and therefore the value of H_{c2} determined in this paper are relevant. On the other hand, we have not been able to detect unambiguously the value of H_{c1}.

Contrary to the transition from the normal to the superconducting state due to temperature, where α is a direct measure of the number of normal electrons left as compared with their number at the critical temperature, this is not true in the magnetic field; the number of fluxoids and their arrangement may considerably influence the value of α. Nevertheless a given induction B corresponds to a certain regular array of flux lines, except in the instability region around $B = 0$. The same applies to the attenuation α, which is thus a singlevalued function of the induction B. This point of view has recently been supported by Forgan and Gough [11], who have found, that the hysteresis almost completely diappears if α is measured as a function of B instead of H_e; this result proves that α reflects the inherent structure of the mixed state. The explanation of a delayed penetration of fluxoids into the sample below H_{c2} offered in [11] as being due to a local variation of H_{c2} in an unhomogeneous sample seems to be rather formal, as the same argument would apply to the increasing magnetic field. It is obvious from our measurements that fluxoids can easily be pushed out with a field, but their penetration is rather difficult, in accordance with the strong position asymmetry of the free energy of a flux line found theoretically by Takács [13]. Favourable sites on the surface may enhance and unfavourable sites may hinder this process. Some analogy with the dislocation processes may be recalled here.

V. CONCLUSIONS

One can conclude that in dirty niobium the attenuation of ultrasound waves yields information concerning the true bulk condition of the mixed state, accountable for in terms of current theories. Even in a sample of unfavourable shape the value of H_{c2} can be determined while the field is increased. The lowering of the temperature and the increasing of the field change rate $\mathrm{d}H/\mathrm{d}t$ enhance the appearance of a sudden step-wise change of fluxoid population, which mostly decrease it.

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REFERENCES

[1] Abrikosov, A. A.: ŽETF *32* (1957), 1442; J. Phys. Chem. Solids *2* (1957), 199.
[2] Gottlieb, M., Garbuny, M., Jones, C.: in *Physical Acoustics*, Vol. 7. Editors W. P. Mason and R. N. Thurston, Academic Press, New York—London 1970.
[3] Bean, C. P., Livingston, J. D.: Phys. Rev. Lett. *12* (1964), 14; De Blois, R. W., De Sorbo, W.: Phys. Rev. Lett. *12* (1964), 499.
[4] Campbell, A. M., Evets, J. E.: Advances in Phys. *21* (1972), 199.
[5] Gottlieb, M., Garbuny, M., Jones, C.: Phys. Lett. *28 A* (1968), 148.
[6] Cleary, R. M.: Phys. Rev. *175* (1968), 587; Forgan, E. M., Gough, C. E.: Phys. Lett. *21* (1966), 133.
[7] Schulze, K., Fuss, J., Schultz, H., Hoffmann, S.: Z. f. Metallkunde *67* (1976), 737; Schulze, K., Jehn, H.: Z. f. Metallkunde *68* (1977), 654.
[8] Brož, J.: *Základy magnetických měření.* NČSAV, Praha 1953; Bozorth, J. M., Chapin, D. M.: J. Appl. Phys. *13* (1942), 320.
[9] Lee, M. J., Dobbs, E. R.: Phys. Lett. *45 A* (1973), 39.
[10] Takács, S.: Czech. J. Phys. B *25* (1975), 1155; B *27* (1977), 336.
[11] Forgan, E. M., Gough, C. E.: Phys. Lett. *26 A* (1968), 602.
[12] Forgan, E. M., Gough, C. E.: J. Phys. F. — Metal Phys. *8* (1978), 1073.
[13] Takács, S.: Z. Phys. *199* (1967), 495; Phys. Stat. Sol. *21* (1967), 709.

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