ANISOTROPY AND TEMPERATURE DEPENDENCE OF LOWER AND UPPER CRITICAL FIELD IN TETRAGONAL SUPERCONDUCTOR In_3Sn

By W. Zacharko and R. Kubiak

Institute for Low Temperature and Structure Research, Polish Academy of Sciences, Wrocław*

(*Received November 16, 1979*)

The anisotropy of the lower H_{c1} and the upper H_{c2} critical fields of tetragonal intrinsic type II superconductor was studied for the first time. The temperature and crystal orientation dependences of both critical fields were measured for the two single crystals of In_3Sn by using the integration method, and anisotropic parameters were determined by means of some theoretical models.

1. *Introduction*

The anisotropy of the critical fields in superconductors with a cubic lattice has been subject of intensive investigations in recent years [1–5]. A few experiments to determine the H_{c2} anisotropy were carried out for the type II superconductors with hexagonal lattice [6–8]. The anisotropy of the H_{c2} in layered tetragonal superconductors with quasi two-dimensional nature of the electron system has been investigated in [9, 10]. However, the anisotropy of the lower H_{c1} and the upper H_{c2} critical fields in the intrinsic type II superconductors with the tetragonal lattice has not been investigated yet.

The intermetallic compounds are good materials to study the pure-limit behaviour of the intrinsic type II superconductors [11, 12]. The ratio between the coherence length ξ and the mean-free path l in the superconducting intermetallic compounds has a large value which is very close to the pure limit. The presence of the intermediate β and γ phases is a characteristic feature of the In-Sn system. The lattice structure of the β-phase is found to be face-centered tetragonal [13]. In the present paper we report the measurements of the temperature dependence and anisotropy of the lower H_{c1} and the upper H_{c2} critical fields in the single crystal In_3Sn. The superconducting and electronic (anisotropic) fundamental parameters are determined and the comparison between the experimental data and theory is given in the framework of some theoretical models.

* Address: Instytut Niskich Temperatur i Badań Strukturalnych, Polska Akademia Nauk, 53-529 Wrocław, Próchnika 95, Poland.

2. *Experimental procedure*

The samples were prepared by the thermal synthesis method. Appropriate amounts of indium and tin 5N purity were mixed in stoichiometric ratio and melted in vacuum. A small droplets of the liquid metals were then cooled down slightly below the peritectic point. The samples were kept at the temperature for several days and then slowly cooled down to the room temperature. The phase β may be cooled to the liquid helium temperature by quenching [14, 15]. Eventually, slowly decreasing temperature leads to the decomposition of β-phase and the destruction of monocrystal In_3Sn [15, 16]. The internal structure of the specimens was checked by X-rays which confirmed the presence of only the β-phase monocrystal. The measurements were performed in an automatic single-crystal diffractometer with the graphite monochromated molybdenum radiation. The crystallographic directions have been determined by X-rays with an accuracy of 1°. The values of the critical fields have been estimated from the magnetization curves obtained by using the integration method [17]. The sample was fixed inside a pick-up coil which was connected in series opposition with a similar bucking coil. The external magnetic field was changed continuously. The difference in the terminal voltages of the both coils, after being amplified in a d.c. amplifier, was integrated in an electronic integrator. This integrated voltage is proportional to the magnetic moment of the specimen and can be plotted continuously versus the applied magnetic field by *X-Y* recorder. The complete coils and the sample was in a bath of the liquid helium inside inner helium dewar. The temperature below 4.2 K was obtained by reducing the vapour pressure of the liquid helium and was stabilized by means of the diaphragm manostat. The temperature of the sample was measured by a resistance germanium thermometer. The temperatures were kept constant at certain values with ± 0.01 degree of the deviations and measured with an accuracy of 0.005 K. The magnetic field produced by a superconducting magnet was homogeneous to within 0.1% over the sample volume. The superconducting magnet was immersed in a bath of liquid helium at atmospheric pressure inside outer helium dewar. The field sweep rate was kept to below 20 Oe/s. The resistivity of each sample was measured at the room temperature and at 4.2 K in the magnetic field about 5 kOe by a mutual inductance technique [18].

3. *Results*

The two single crystalline samples of the spherical shape with about 3 mm in diameter were studied. The residual resistance ratio $R_{300}/R_{4/2} \approx 30$ have indicated that the single crystals were a good quality. The magnetization curves of the In_3Sn were carried out in an external magnetic field which could be parallel to arbitrary crystallographic direction because of the spherical shape of the sample. Our measurements were made in the magnetic field parallel to the [100] and [001] crystallographic direction, respectively. The isothermal magnetization curve as a function of the magnetic field was registered directly by the *X-Y* recorder in increasing and decreasing magnetic field at a chosen temperatures from the interval between 1.3 K and 4.2 K. A typical example of the magnetization curves in the [100] direction for the sample of the In_3Sn is presented in Fig. 1. It can be seen from Fig. 1 that the magnetization curves have a small hysteresis and a frozen-in flux. The lower critical field H_{c1} and the upper critical field H_{c2} were determined from these curves. The dependence of the critical fields on the reduced temperature for the crystallographic direction [100] and

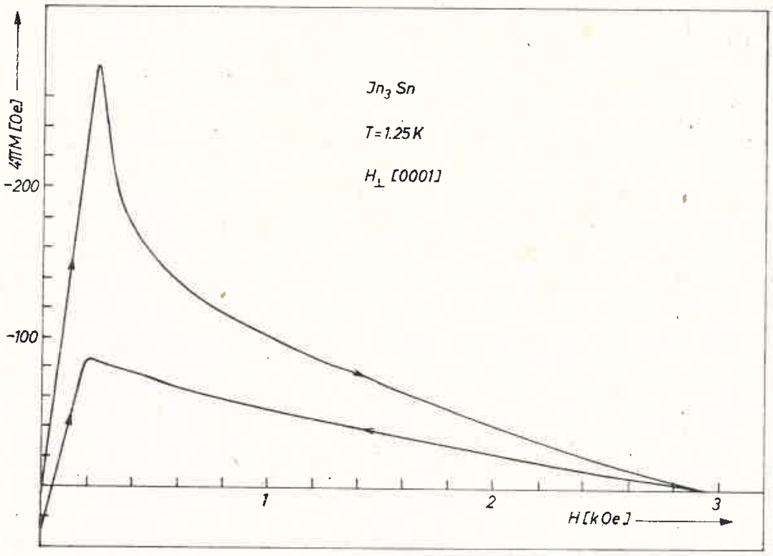

Fig. 1. The typical magnetization curve of In_3Sn at 1.30 K

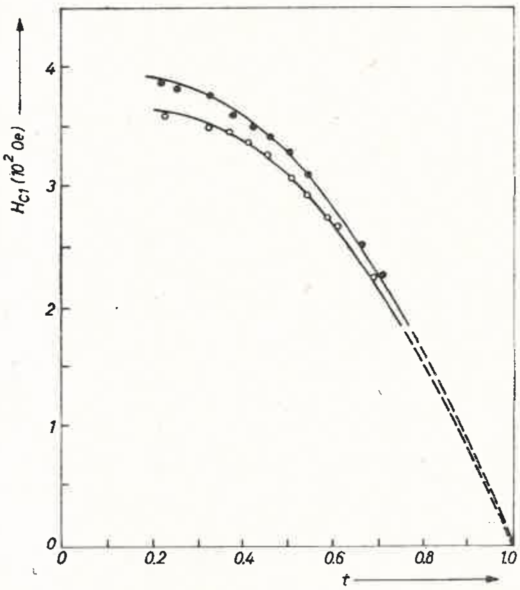

Fig. 2. The dependence of H_{c1} versus reduced temperature t (where ○ and ● denote $H \perp c$ and $H \| c$, respectively)

[001] is presented in Fig. 2 and Fig. 3. The experimental curves from Figs. 2 and 3 indicate the presence of a quite significant anisotropy of the upper and lower critical field. The anisotropy effects of an uniaxial type II superconductors have been studied in the framework the of Ginzburg-Landau theory.

Hence, the upper critical field H_{c2} depends on the direction of the magnetic field with respect to the crystallographic c-axis [19]. It is well-known that by using the effective

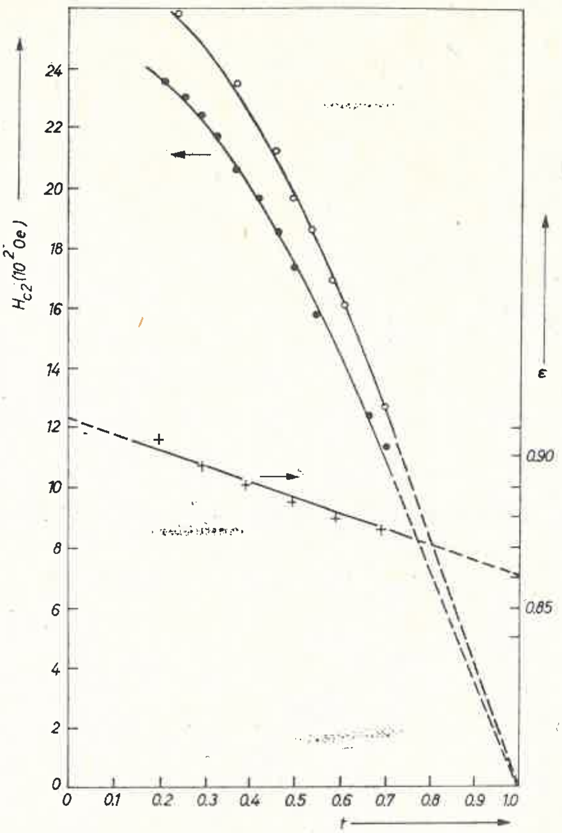

Fig. 3. The dependence of H_{c2} and ε versus reduced temperature t (where ○, ● and + denote $H \perp c$, $H \| c$ and anisotropy mass parameter, respectively)

mass approximation for the case of the uniaxial type II superconductors [9] the mass anisotropy parameter ε can be defined as follows

$$\varepsilon = (m_\perp / m_{\|})^{1/2} = H_{c2\|}/H_{c2\perp} = \xi_{\|}/\xi_\perp, \tag{1}$$

where the effective mass $m_\perp$, $m_{\|}$ and the coherence length $\xi_\perp$, $\xi_{\|}$ are chosen in the perpendicular or parallel direction to the c-axis, respectively. The value of the parameter ε for In_3Sn is given in Table I while its temperature dependence is presented in Fig. 3. Subsequently, by using the following relation

$$H_{c2}(t) = H_{c2}(0)\,(1 - at^2 + bt^4), \tag{2}$$

where a and b are arbitrary constants and t denotes the reduced temperature $t = T/T_c$, the values of the critical field $H_{c2}(0)$ from Table I were obtained by extrapolation.

The coherence lengths can be expressed in the following way

$$\xi_{0\perp}^2(0) = \Phi_0/2\pi H_{c2\parallel}(0), \quad \xi_{0\perp}(0)\xi_{0\parallel}(0) = \Phi_0/2\pi H_{c2\perp}(0), \tag{3}$$

where Φ_0 is the flux quantum. Hence, the coherence lengths $\xi_{0\parallel}(0)$ and $\xi_{0\perp}(0)$ at zero temperature $T = 0$ K are determined and the respective values for parallel and perpendicular critical fields are given in Table I. Finally, the reduced field gradient and the normalized field can be determined from the relations

TABLE I

Fundamental parameters of In_3Sn

Parameters \ Direction of H	$H \perp c$ [100]		$H \parallel c$ [001]
T_c [K] [14]		5.95	
		0.862	
$H_{c2}(0)$ [Oe]	2830		2593
$H'_{c2}(1)$ [Oe]	−4216		−3700
$h^*(0)$	0.67		0.70
$\langle v_F^2 \rangle^{1/2}$ [10^7 cm/s]	1.63		0.74
$\xi_0(0)$ [Å]	425		390

$$h^*(0) = H_{c2}(0)/H'_{c2}(1), \quad H'_{c2}(1) = (dH_{c2}/dt)_{t=1}, \tag{4}$$

respectively.

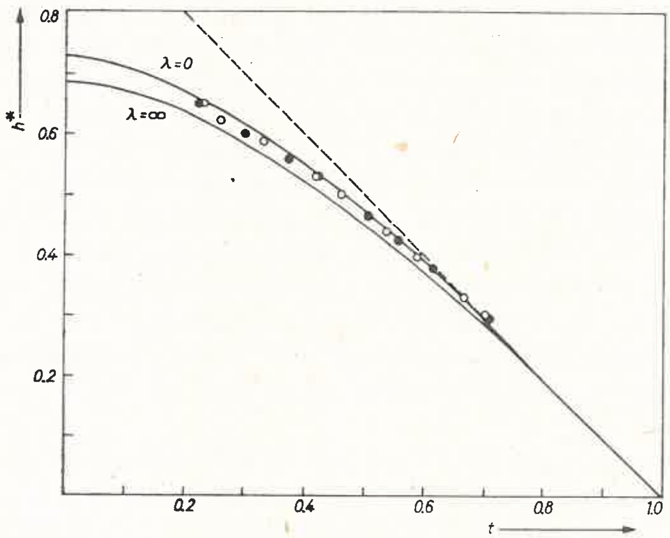

Fig. 4. The normalized field h^* versus reduced temperature t. Solid lines denote two limiting cases of $h^*(t)$ for $\lambda = 0$ (pure limit) and $\lambda = \infty$ (dirty limit). Dashed line marks the limiting slope (equal to −1) of $h^*(t)$ at $t = 1$. The signs ○ and ● are our experimental points for $H \perp c$ and $H \parallel c$ in In_3Sn, respectively

Above quantities for In_3Sn are presented in Table I and the relation $h^*(t)$ is depicted in Fig. 4. The values of $h^*(t)$ at low temperature are smaller than the ones which can be calculated theoretically in the limits of pure and dirty type II superconductors [20].

It seems to us that a weak electron-phonon coupling in In_3Sn is likely to be responsible for this fact. The well-known relation between velocity at the Fermi surface $\langle v_F \rangle$, $H'_{c2}(1)$ and T_c in the case of the type II superconductors with a weak electron-phonon coupling was determined by Werthamer and McMillan in [21]

$$H'_{c2}(1) = -\frac{6c(2\pi k T_c)^2}{7e\hbar\zeta(3)\langle v_F^2 \rangle}, \tag{5}$$

where c is the light velocity, k denotes the Boltzmann constant, $\hbar = h/2\pi$ is the reduced Planck constant, e denotes the electron charge and $\zeta(3) = 1.202$. The parameter $\langle v_F^2 \rangle^{1/2}$ for two crystallographic directions can be calculated from Eqs. (4) and (5) and its values are presented in Table I.

4. *Discussion*

The fundamental parameters to describe the anisotropy behaviour of the type II superconductors have been given in the previous section. By taking into account Eqs. (1) and (2) together with parameters a and b (Eq. (2)) taken from experimental data, a small increase of the mass anisotropy parameter ε with decreasing temperature can be easily seen from Fig. 3. On the other hand, the parameter ε should be temperature independent in the framework of the anisotropic Ginzburg-Landau theory. Then, its weak temperature dependence should result from nonlocal effects while the value from the extrapolation to the critical temperature T_c is the following: $\varepsilon = 0.86$ (see Fig. 3). The same weak temperature dependence of ε was found by Kostorz et al. [6] and Zacharko et al. [7]. However, it is obvious that the simple effective mass model seems to be insufficient for the description of the temperature dependence of the H_{c2} anisotropy in the type II uniaxial superconductors without impurities. Besides, a slow increase of the lower critical field H_{c1} anisotropy is observed with increasing temperature in In_3Sn. In this case, the ratio $H_{c1\parallel}/H_{c1\perp}$ increases from 1.06 to 1.08 in the reduced temperature range $0.3 < t < 0.7$, respectively.

Moreover, it was determined from experimental data that the ratio of the magnetization curve slope in the intermediate mixed state (it describes a regular structure with the regions in mixed state and Meissner state) to its slope in Meissner state was approximately equal to 1.5. This ratio is quite far from the value of 2 which is expected under the assumption that the phase transition is first order for $H = H_{c1}$ (in so-called type II/1 superconductor).

Generally, the thermodynamic critical field H_c is independent on the crystallographic direction hence the area under the magnetization curve should be constant apart from the direction of the external magnetic field with respect to the c-axis. By taking into account the well-known relation

$$2H_c^2 \approx H_{c1}H_{c2} \tag{6}$$

it is easy to calculate for example H_{c1} anisotropy from the H_{c2} anisotropy, which is taken from experimental data, under the assumption that H_c is independent of the crystallographic direction. In our case the ratio $H(t)_{c\perp}/H_{c\|}(t)$ for the reduced temperature range $0.3 < t < 0.7$ changes from 1.01 to 1.03 and it confirms in some ways the relation (6) between H_{c2} and H_{c1} anisotropy under condition that the magnetization is continuous at H_{c1}. A similar dependence between both critical field anisotropies in the type II superconductors with a cubic lattice has been obtained in [4–5]. Further experimental investigations are required for stating what kind of mechanism is responsible for anisotropy of critical fields in the tetragonal superconductors.

The authors are grateful to Professor G. Kozłowski for helpful discussions.

REFERENCES

[1] S. J. Williamson, *Phys. Rev.* **B2**, 3545 (1970).
[2] E. Seidl, H. W. Weber, H. Teichler, *J. Low Temp. Phys.* **30**, 237 (1978).
[3] H. W. Weber, E. Seidl, G. Kirshenhofer, *Solid State Commun.* **26**, 795 (1978).
[4] D. K. Christen, P. Thorel, *Phys. Rev. Lett.* **42**, 191 (1979).
[5] H. Kiesing, U. Essmann, W. Wiethaup, *Phys. Rev. Lett.* **71A**, 467 (1979).
[6] G. Kostorz, L. L. Isaacs, R. L. Panosh, C. C. Koch, *Phys. Rev. Lett.* **27**, 304 (1971).
[7] W. Zacharko, R. Kubiak, J. Mazur, *Phys. Status Solidi* (b) **76**, K1 (1976).
[8] M. R. Skokan, R. C. Morris, W. G. Moulton, *Phys. Rev.* **B13**, 1077 (1976).
[9] J. A. Woollam, R. B. Somoano, *Phys. Rev.* **B13**, 3843 (1976).
[10] K. Onabe, M. Naito, S. Tanaka, *J. Phys. Soc. Jap.* **45**, 50 (1978).
[11] H. Ullmaier, R. H. Kernohan, E. Cruceanu, E. Hering, *J. Low Temp. Phys.* **3**, 71 (1971).
[12] H. Ullmaier, E. Cruceanu, *Rev. Roum. Phys.* **17**, 547 (1972).
[13] C. G. Fink, E. R. Jette, S. Katz, F. J. Schnettler, *Trans. Electrochem. Soc.* **88**, 229 (1945).
[14] W. Zacharko, J. Mazur, *Acta Phys. Pol.* **A46**, 109 (1974).
[15] R. Kubiak, M. Wołcyrz, W. Zacharko, *J. Less. Common Met.* **65**, 263 (1979).
[16] S. F. Bartram, W. G. Moffatt, B. W. Roberts, *J. Less. Common Met.* **69**, 9 (1971).
[17] W. A. Fietz, *Rev. Sci. Instrum.* **36**, 1621 (1965).
[18] M. D. Daybell, *Rev. Sci. Instrum.* **38**, 1412 (1967).
[19] T. Takanaka, H. Ebisawa, *Progr. Theor. Phys.* **47**, 1718 (1972).
[20] E. Helfand, N. R. Werthamer, *Phys. Rev.* **147**, 288 (1966).
[21] N. R. Werthamer, W. L. McMillan, *Phys. Rev.* **158**, 415 (1967).