Magnetic Behavior of Type-II Superconductors with Small Ginzburg–Landau Parameters

J. Auer and H. Ullmaier

Institut für Festkörperforschung der Kernforschungsanlage, 517 Jülich, Germany

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Type-II superconductors with $\kappa \approx 1$ exhibit a first-order phase transition at H_{c1} , which is due to an attractive interaction between flux lines. This behavior shows up as a discontinuous increase of the flux density from zero to a certain value B_0 . We have measured B_0 as a function of temperature T, Ginzburg-Landau parameter κ , and impurity parameter $\alpha = 0.882 \xi_0/l$, and have determined the critical values of these parameters below which a first-order transition occurs at H_{c1} . We further investigated the dependence of the parameters κ_1 and κ_2 on l and T, and we show that only the condition $\kappa_1(T) = 1/\sqrt{2}$ determines the transition from type-I to type-II behavior. Comparison of our experimental results with recent theoretical treatments shows only qualitative agreement.

I. INTRODUCTION

The magnetic properties of type-II superconductors with large Ginzburg-Landau parameters κ \gg 1 are in good agreement with theory. For lower- κ values the quantitative agreement between theory and experiment begins to fail.¹ For materials with $\kappa \approx 1$, the predictions of earlier theories become even qualitatively wrong. The most striking discrepancy is a discontinuous increase of the flux density from zero to a certain value B_0 when the lower critical field H_{c1} is exceeded. Although this behavior was recognized several years ago,² no explanation was given until 1970, when Krägeloh³ observed an intermediate mixed state in a low- κ type-II superconductor using the decoration technique of Essmann and Träuble.⁴ From his observations he concluded that an attractive interaction exists between flux lines. This has been verified recently by neutron-diffraction experiments in pure Nb. 5 If the flux lines attract each other above the lower critical field H_{c1} , flux will enter the sample volume until a lattice with a lattice parameter d_0 is formed, where d_0 is the distance between flux lines, where the attractive interaction changes to a repulsive one. The sudden change of flux gives rise to a jump B_0 , in the flux density. B_0 is related to d_0 by

$$B_0 = \frac{2\phi_0}{\sqrt{3}d_0^2} , \qquad (1)$$

where ϕ is the flux quantum.

It is therefore possible to determine the "equilibrium" lattice parameter d_0 by simple magnetization measurements⁶ as first done by Kumpf⁷ for Pb-Tl alloys. We have made similar measurements on TaN and NbN systems for a wide range of Ginzburg-Landau parameters κ and electron mean free paths l, and for different temperatures T. The alloys TaN and NbN were prepared so that the magnetization curves were almost completely reversible. This enabled us to determine relevant quantities (e.g., B_0 , κ , the thermodynamical critical field H_c , etc.) with high precision. These results are presented in Sec. IIIC. We have also investigated the behavior of the generalized Ginzburg-Landau parameters κ_1 and κ_2 , especially in the neighborhood of the transition from type-I to type-II superconductivity (Sec. IIIB). In order to ensure that the observed effects are simply and solely due to changes of secondary superconducting properties like κ and l, we also measured H_c and T_c . It was found that these primary properties are only negligibly altered by the addition of nitrogen for the concentration range investigated (Sec. IIIA). The first theoretical indication for an attractive interaction between flux lines can be found in calculations by Eilenberger and Büttner,⁸ who obtained oscillations in the magnetic field distributions of an isolated flux line as soon as κ was decreased below a certain value. A similar field reversal⁹ was obtained by Halbritter¹⁰ and Dichtel.¹¹ Whereas these considerations based on a nonlocal relation between current density and vector potential of the magnetic field are probably important for low temperatures, Jacobs^{12,13} showed that an attractive interaction energy can also be expected close to the transition temperature. Jacobs's calculations are based on the Neumann-Tewordt extension¹⁴ of the Ginzburg-Landau theory. A comparison of the experimental results with the new theoretical attempts (Sec. IIIE) shows that at present the magnetic behavior of $low - \kappa$ type-II superconductors is in principle understood, but there is still need for better quantitative agreement.

II. EXPERIMENTAL

A. Starting Materials

The starting materials for our samples were Ta and Nb for the following reasons. (a) The

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transition temperatures are in a convenient range for experiments. (b) The superconducting parameters of both materials are well known. (c) Pinning centers which are usually most efficient close to H_{c1} must not be present in the samples because they could mask the effects which will be observed. Nb and Ta dissolve relatively large amounts of nitrogen, and the statistically distributed N interstitials are not effective for pinning, but only reduce the electron mean free path while increasing κ in the desired way. (d) The Ginzburg-Landau parameter κ_0 of pure Ta is smaller than $1/\sqrt{2}$. Controlled additions of N yield samples with κ $\approx 1/\sqrt{2}$ in which the change from type-I to type-II behavior can be studied. Pure Nb has $\kappa_0 > 1\sqrt{2}$; i.e., the behavior of a pure type-II superconductor can be investigated (this case is mostly treated by theory).

The starting materials used had a purity of 99.996%, the main impurities being Ta in Nb and vice versa. Samples were polycrystalline wires of 1.25-mm diameter and 60-mm length. After outgassing the Ta and Nb at 2600 and 2300 °C, respectively, in a vacuum of $3-8 \times 10^{-11}$ Torr for 3 h, resistivity ratios as measured at 300 and 4.2 K were between 1500 and 2000 for the respective metals. Then nitrogen was introduced by heating samples to 1800 °C in a nitrogen atmosphere from 10^{-6} to 10^{-4} Torr in accordance with the amount of nitrogen desired. The maximum concentrations of nitrogen were 0.38 at.% in Ta and 0.07 at.% in Nb, as determined by resistivity increases of 1800 and 360 n Ω cm, respectively. A final annealing treatment was given to the wires by heating in air at 400 °C for 2 min. This procedure creates a strong concentration gradient of oxygen and nitrogen in a surface layer of about $1-\mu m$ thickness. It is known^{15,16} that this type of surface strongly decreases surface barriers for flux lines. After this treatment samples exhibited almost completely reversible magnetization curves (see Fig. 1). Bulk properties of samples remained unchanged by the surface treatment. This was confirmed by showing (i) that the residual resistivity and the upper critical field before and after the oxidation were the same, and (ii) that the T_c and H_c values of the pure samples were in excellent agreement with values given in the literature. $^{17,\,18}$ Results are compiled in Table I for all samples together with some characteristic properties.

B. Magnetization and Resistivity Measurements

The magnetization curves were measured by slowly sweeping the external field H from zero to a value above the upper critical field H_{c2} and back to zero while integrating the voltage generated in a coil system that encircled a sample. The coil system consisted of a pickup coil and a compensation coil to cancel out the signal produced in the space between the sample and the pickup coil. The external field was supplied by a superconducting sixth-order solenoid and was constant within $\pm 0.2\%$ over the sample length. Special care was taken to ensure that the sample axis was parallel to H. Temperatures below 4.2 K were achieved by reducing the pressure above the liquid

| | | | , . | Ginzburg-Landau |
|----------|-------------------------------|----------------|----------------------------|-----------------|
| a . | Residual resistivity | Critical temp. | Impurity parameter | parameter |
| Specimen | $\rho_0 (n\Omega \text{ cm})$ | T_{c} (°K) | $\alpha = 0.882 \xi_0 / l$ | К |
| Та | 7.1 | 4.48 | 0.0098 | 0.355 |
| TaN3 | 163 | 4.44 | 0.225 | 0.453 |
| TaN 5 | 263 | 4.43 | 0.364 | 0.513 |
| TaN 13 | 305 | 4.42 | 0.420 | 0.538 |
| TaN 4 | 318 | 4.42 | 0.439 | 0.546 |
| TaN 8 | 367 | 4.41 | 0.506 | 0.577 |
| TaN 9 | 393 | 4.40 | 0.543 | 0.591 |
| TaN17 | 400 | 4.40 | 0.552 | 0.595 |
| TaN 19 | 417 | 4.40 | 0.576 | 0.605 |
| TaN 20 | 434 | 4.40 | 0.600 | 0.615 |
| TaN 6 | 524 | 4.38 | 0.724 | 0.670 |
| TaN18 | 585 | 4.37 | 0.810 | 0.705 |
| TaN14 | 640 | 4.36 | 0.884 | 0.735 |
| TaN 12 | 660 | 4.35 | 0.910 | 0.751 |
| TaN 7 | 666 | 4.35 | 0.920 | 0.755 |
| TaN 2a | 815 | 4.34 | 1.125 | 0.845 |
| TaN1 | 1967 | 4.22 | 2.710 | 1,535 |
| Nb 1 | 7.96 | 9.25 | 0.0056 | 0.78 |
| Nb 2 | 18.1 | 9.25 | 0.0133 | 0.792 |
| Nb 3 | 70.6 | 9.24 | 0.052 | 0.824 |
| Nb 4 | 360 | 9.22 | 0.265 | 1.03 |

TABLE I. Parameters which characterize the superconducting properties of TaN and NbN samples: ρ_0 , residual resistivity; T_c , transition temperature; α , impurity parameter; and κ , Ginzburg-Landau parameter.

helium. Measurements above 4.2 K were performed in a heated container filled with 1-Torr He gas and the sample temperature was maintained within 0.005 K of the desired value by an electronic controller.

The electrical resistivity ρ_0 was measured by a conventional four-probe technique at 4.2 K. Fields larger than H_{c3} were applied to convert samples to the normal state. No magnetoresistance was detectable for the *Nb*N samples, but for the pure Nb samples ρ_0 was obtained by a quadratic extrapolation to H = 0. No magnetoresistance was observed for the Ta and *Ta*N samples because here H_{c3} at 4.2 K is very small (<50 Oe).

III. RESULTS AND DISCUSSION

A. Primary Superconducting Properties

The thermodynamic critical field H_c of a superconductor can be obtained from its equilibrium magnetization curve M(H) by

$$\frac{H_c^2}{8\pi} = -\int_0^{H_c 2} M(H) dH \quad .$$
 (2)

Since the magnetization curves of our samples were reversible except for a very small region at H_{c1} , we were able to deduce H_c from Eq. (2) without additional assumptions concerning surface currents, pinning effects, etc. The H_c values obtained in this way followed a parabolic temperature dependence very closely. Extrapolation of this parabolic function to T = 0 yields $H_c(0)$. The transition temperature T_c was obtained by extrapolating the $H_c(T)$ values to $H_c = 0$. Values for the thermodynamic critical field at zero temperature and for the transition temperature are shown in Fig. 2 for TaN samples as a function of the residual resistivity ρ_0 . The slight decrease of $H_c(0)$ and T_c with increasing nitrogen content is probably due to the reduction of the anisotropy of the energy gap.¹⁹ This is indicated by the fact that the ratio $[H_c(0)/T_c]^2$, which is proportional to the density of states N(0) at the Fermi level, stays constant. This means that N(0) is not noticeably changed by the addition of small amounts of nitrogen. We may therefore conclude that all samples in our systems TaN have almost identical primary superconducting properties; i.e., the only electronic parameter which is changing from sample to sample is the electron mean free path l. The same is true for our NbN samples, where the maximum nitrogen concentration is even smaller than in TaN.

The knowledge of the temperature dependence of H_c is useful for another reason. It allows us to calculate the coherence length ξ_0 and the London penetration depth $\lambda_L(0)$ for pure superconductors. Near the transition temperature H_c is given by²⁰

$$H_{c}(t) = \frac{\varphi_{0}}{2\pi\sqrt{2}\,\xi(t)\lambda(t)} = \frac{-\varphi_{0}\kappa}{2\pi\sqrt{2}\,\lambda(t)^{2}}; \qquad (3)$$

 ξ is the Ginzburg-Landau coherence length, λ the penetration depth, and *t* the reduced temperature.



FIG. 2. Values of critical temperature T_c and thermodynamical critical field at T=0, H_c (0), for TaN samples are given as a function of their residual resistivity.

With

$$\lambda(t) = \frac{1}{\sqrt{2}} \lambda_L(0)(1-t)^{-1/2}$$
(4)

we obtain

$$\lambda_L(0) = \left(\frac{\pi\sqrt{2}}{\varphi_0\kappa} \left| \frac{dH_c}{dt} \right| \right)^{-1/2} \quad , \tag{5}$$

and because for the pure limit

$$\kappa = \frac{0.96\lambda_L(0)}{\xi_0} , \qquad (5a)$$

we can also determine ξ_0 if κ is known. With $\kappa = 0.355$ for Ta (see Sec. III A), we obtain $\lambda_L(0) = 345$ Å and $\xi_0 = 925$ Å in good agreement with Ref. 21. For Nb ($\kappa = 0.78$) we obtain $\lambda_L(0) = 315$ Å and $\xi_0 = 390$ Å, which is somewhat lower than values in the literature.¹⁸ We believe that an estimate of the characteristic lengths by Eqs. (5) and (5a) is more reliable than using expressions valid for T = 0, as done in Ref. 18, because nonlocal effects which have a large influence at low temperatures are negligible near T_c .²²

B. Dependence of κ_1 and κ_2 on Temperature and Electron Mean Free Path

The Ginzburg-Landau-Maki parameters κ_1 and κ_2^{23} at an arbitrary temperature follow from a generalization of two relations defining the Ginz-burg-Landau parameter κ :

$$\kappa_{1}(T) = \frac{H_{c2}(T)}{\sqrt{2} H_{c}(T)}$$
(6)

and

$$\kappa_{2}(T) = \left(\frac{1}{8\pi \, dM/dH} \Big|_{H_{c2}} \frac{1}{1.16} + \frac{1}{2}\right)^{1/2} \quad . \tag{7}$$

In the limit as $T \rightarrow T_c$, both definitions yield the

same value, i.e., the Ginzburg-Landau parameter κ . Numerical calculations²⁴ show that for temperatures $T < T_c$ the inequality $\kappa_2 > \kappa_1 > \kappa$ holds. Since in our measurements on TaN samples, which cover a wide range of mean free paths, the critical fields H_c and H_{c2} and the slope of the magnetization curve near H_{c2} could be measured very accurately, it seems worthwhile to compare the results with theoretical calculations.

In Fig. 3 the parameters κ_1 and κ_2 are shown as a function of temperature for different TaN samples characterized by their specific residual resistivity. Comparison between κ_1 and κ_2 values of each sample shows that at any temperature κ_2 is larger than κ_1 , while the change from type-I to type-II behavior occurs at the temperature where $H_{c2}=H_c$, i.e., $\kappa_1=1/\sqrt{2}$. Since κ_2 is larger than κ_1 , that means that the "tail" in the magnetization curve above H_{c1} vanishes without the slope dM/ $dH|_{H_{22}}$ going to infinity. This could probably be confirmed by supercooling measurements. In the diagrams of Fig. 4 the parameters κ_1 and κ_2 normalized to the Ginzburg-Landau parameter κ are plotted as a function of the reduced temperature. For three samples in this figure, values are given for theoretically calculated values of κ_1/κ and κ_2/κ for the corresponding ratio ξ_0/l assuming pure s scattering $(l_{tr}/l=1)$.²⁴ ξ_0/l was calculated using the ξ_0 values determined in Sec. IIIA. lwas found as the ratio of the known $(\rho_0 l)$ values for Ta²⁵ and Nb²⁶ and the measured resistivity. Obviously, the observed temperature dependence of κ_1 is much stronger than predicted by theory. The discrepancy between theoretical and experimental values is usually attributed to the neglect of Fermisurface anisotropy in the theoretical treatment.²⁷ This assumption is supported by the fact that the best agreement is obtained when the mean free



FIG. 3. Generalized Ginzburg-Landau parameters κ_1 and κ_2 are plotted in (a) and (b), respectively, as a function of the reduced temperature t for different TaN samples. Each curve is identified by its residual resistivity ρ_0 . Extrapolation of κ_1 (t) and κ_2 (t) to $t \rightarrow 1$ yields the same Ginzburg-Landau parameter κ for a given ρ_0 .

path *l* is very short (see Fig. 4), i.e., the more the anisotropy of the energy gap is smeared out by scattering. It is not yet clear why the agreement of κ_2 with the theoretical prediction is so good, whereas κ_1 deviates, since the simplifying assumptions of the theory²⁴ are the same for both parameters.

It was found experimentally that the Ginzburg– Landau parameter, as well as the parameters κ_1 and κ_2 , at all temperatures investigated follows a linear dependence on the residual resistivity very closely, as shown in Fig. 5.

For the Ginzburg–Landau parameter κ the experimental data fit the equation

$$\kappa = \kappa_0 + 0.60\rho_0 \quad , \tag{8}$$

where ρ_0 is given in $\mu \Omega$ cm. Inserting literature values for the coefficient of the electronic specific heat for Ta^{17,28} ($\gamma = 0.66 \text{ erg cm}^{-3} \,^{\circ}\text{K}^{-2}$) into the theoretical expression of Goodman²⁹ we obtain

$$\kappa = \kappa_0 + 7.5 \times 10^{-3} \gamma^{1/2} \rho_0 = \kappa_0 + 0.61 \rho_0 \quad , \tag{8a}$$

in excellent agreement with Eq. (8). A linear extrapolation of the measured κ values to $\rho_0 \rightarrow 0$ yields the Ginzburg–Landau parameter κ_0 for pure tantalum: $\kappa_0 = 0.355 \pm 0.010$. This value is rather low compared with extrapolations from other Ta-alloy systems, ²⁸ but agrees very well with super-cooling measurements on pure Ta, which give a value $\kappa_0 = 0.34 \pm 0.01$.²¹

C. Discontinuity of Flux Density at H_{c1}

As already outlined in the Introduction, the attractive interaction between flux lines in $low-\kappa$ type-II superconductors causes a discontinuous increase in the flux density at H_{c1} . To investigate this phenomenon systematically a series of magnetization measurements at various temperatures were performed on Ta and Nb samples whose electron mean free paths were varied by adding different amounts of nitrogen. As a typical example Fig. 1 shows magnetization curves of a TaN sample for which the Ginzburg-Landau parameter was slightly below $1/\sqrt{2}$; i.e., type-I superconductivity was observed near T_c . Due to the temperature dependence of κ_1 , the sample showed type-II behavior at lower temperatures $(T < 0.9T_c)$. In the type-II region a very sharp discontinuity in the flux density appears in both the increasing and decreasing field branches of the magnetization curve. Since the B_0 values obtained from these two curves are slightly different (~ 3%; see Fig. 1) and since the cause for this difference is not yet clear, for the following discussion we define B_0 as the average of the values in increasing and decreasing fields.

In Fig. 6 values of B_0 determined in this way are shown as a function of the reduced temperature for different TaN samples. B_0 decreases with in-



FIG. 4. Ratios κ_1/κ and κ_2/κ are plotted vs the reduced temperature t for some typical TaN samples. Calculated curves (Eilenberger, Ref. 24) are included for pure s scattering; i.e., the transport mean free path $l_{\rm tr}$ is equal to the mean free path $l(l_{\rm tr}/l=1)$. For the pure case, $\rho_0=0$, values extrapolated from TaN alloys in the type-II region are shown.

creasing temperature or increasing impurity parameter as expected, since an increase of these parameters reduces the attractive contributions to the flux-line interactions. For samples with $\kappa < 1/\sqrt{2}$ (upper part of Fig. 6) the B_0 curves end at a temperature t_1 where $\kappa_1(t_1) = 1/\sqrt{2}$, i.e., where the sample changes to type-I behavior. For samples with $\kappa > 1/\sqrt{2}$, B_0 can be determined in the whole temperature range. At high temperatures B_0 exhibits a linear dependence on t similar to the other critical fields. Linear extrapolation to $B_0 \rightarrow 0$ shows that the jump in the magnetization vanishes at a temperature $T^* < T_c$. The quantity $T_c - T^*$ increases with increasing Ginzburg-Landau parameter. The curves for the three NbN

samples $(\kappa > 1/\sqrt{2})$ exhibit qualitatively the same behavior.

From the measured B_0 values one can calculate the corresponding flux-line lattice parameter d_0 by Eq. (1) (right-hand scale of Fig. 6). Figure 7 shows d_0 as a function of the reduced temperature for our NbN samples. Figure 7 contains an additional point measured directly by neutron-scattering experiments⁵ which is in good agreement with the values obtained from B_0 .

D. Two Kinds of Type-II Superconductors

The discontinuous increase of the flux density at H_{c1} in low- κ type-II superconductors causes a first-order phase transition, in contrast to the



FIG. 5. Ginzburg-Landau parameter κ and the generalized Ginzburg-Landau parameters κ_1 (t=0.5) and κ_2 (t=0.5) for TaN samples show a linear dependence on the residual resistivity ρ_0 . Extrapolation of κ (ρ_0) to $\rho_0 \rightarrow 0$ yields the Ginzburg-Landau parameter of pure tantalum ($\kappa_0 = 0.355$).

second-order transition observed in materials with large Ginzburg-Landau parameter. For a given temperature t we may therefore distinguish between three kinds of magnetic behavior of superconductors, depending on their κ_1 value: (a) type-I superconductors in the region $0 < \kappa_1(t) < 1/\sqrt{2}$; (b) type-II/1 superconductors with $1/\sqrt{2} < \kappa_1$ (t) $< \kappa_{cr}$ (*t*), which exhibit a first-order transition at H_{c1} ; (c) type-II/2 superconductors with κ_{cr} (t) < κ_1 (t) $<\infty$, which show the "normal" type-II behavior. $\kappa_{\rm cr}(t)$ is the κ_1 value where d_0 becomes infinite, i.e., where the attractive interaction vanishes. κ_{er} depends on temperature t and impurity parameter α and can be determined from our data by extrapolating the B_0 (κ_1) values to $B_0 \rightarrow 0$. For comparison with theory it is appropriate to refer to κ rather than κ_1 . This can easily be done because the relations between $\kappa_1(t)$ and κ are known for each sample (Sec. IIIB). In this way we can construct a "phase diagram" for the magnetic behavior of superconductors with varying Ginzburg-Landau parameters. For the system TaN this is shown in Fig. 8. Four regions can be distinguished in the figure: type-I, type-II/1, and type-II/2 superconductivity for t < 1, and normal conductivity for *t* > 1.

E. Conclusions

Up to now a discontinuity in the magnetization curve has been observed in several low- κ type-II superconductors: *Ta*N and *Nb*N (this work); *Pb*Tl, ⁷ Nb, ^{2,5} and V. ³⁰ From these measurements the distance d_0 can be calculated by using Eq. (1).

The values of d_0 depend on temperature t, Ginzburg-Landau parameter κ , and impurity parameter α of the samples. From the curves of d_0 vs κ , no systematic correlation of the values from different materials can be recognized. However, if one plots the ratio $d_0/\lambda_L(l,t)$ vs κ , as suggested by theory, ^{10,11} a remarkably simple relation is obtained (Fig. 9). Within the experimental uncertainties all $d_0/\lambda_L(l,t)$ values lie on a universal curve and depend (for constant temperature) only on the Ginzburg-Landau parameter of the material. $\lambda_L(l,t)$ is the mean-free-path-dependent London penetration depth given by¹⁰

$$\lambda_L(l, t) = \lambda_L(\infty, t) \left(1 + \frac{\xi_F(\infty, t)}{l} \right)^{1/2} , \qquad (9)$$

where $\lambda_L(\infty, t)$ is the London penetration depth for pure material. At zero temperature $\xi_F(\infty, t)$ is related to the BCS coherence length ξ_0 in the following way: $\xi_F(\infty, 0) = (\frac{1}{2}\pi)\xi_0$. It is evident that the universal relationship between d_0/λ_L and κ for different materials (shown in Fig. 9) is a strong indication that the type-II/1 behavior is a general phenomenon for all low- κ type-II superconductors.

Investigation by decoration technique³ and neutron diffraction⁵ shows that the discontinuity of the flux density B_0 at H_{c1} is due to an attractive interaction between flux lines. As already mentioned in the Introduction, two causes for an attractive interaction are considered theoretically. The first is due to a field reversal in the magnetic field distribution of an isolated flux line which can give rise to magnetic attraction between flux lines. Such a field reversal has been obtained by theoretical calculations^{10, 11, 31} which take into account a nonlocal relationship between current density and the vector potential of the magnetic field. Experi-



mentally a field reversal has been observed by Drangheid and Sommerhalder⁹ in type-I superconductors. Theoretical calculations also predict distances r_0 between the flux-line center and the minimum in the microscopic field. Because the shape of isolated flux lines will be changed when approaching other vortices, the distance r_0/λ_L need not be exactly equal to the "equilibrium" distance d_0/λ_L of the flux-line lattice at H_{c1} . Nevertheless, these two quantities should not differ too much. Comparison between theory^{31,32} and experiment shows that the calculated values r_0/λ_L are about a factor of 1.5-2 larger than our measured values d_0/λ_L . A second possible reason for an attraction between flux lines in a low- κ material arises from an attractive condensation energy term, which may exceed the electromagnetic repulsion. The relatively large magnitude of this attractive condensation energy term occurs because of a more uniform distribution of the superconducting order parameter ψ , caused by the overlap of vortices in the flux-line lattice. Calculations^{12,13} based on the Neumann-Tewordt¹⁴ extensions of the Ginzburg-Landau theory yield for $T < T_c$ a dominance of the attractive term for the interaction energy if κ is below a certain value. The theory is not able to predict any d_0/λ_L values and is only valid close to



FIG. 7. Flux-line lattice parameter d_0 plotted as a function of the reduced temperature for the NbN samples.



FIG. 8. Phase diagram of the magnetic behavior for the TaN system is shown. The Ginzburg-Landau parameter κ (lower abscissa) and the impurity parameter α (upper abscissa) are proportional to the amount of dissolved nitrogen.



FIG. 9. Flux-line lattice parameter d_0 has been normalized by using the London penetration depth $\lambda_L(l, t)$ for constant temperature t=0.3. The magnitude of d_0/λ_L vs the Ginzburg-Landau parameter κ gives values which depend only on the Ginzburg-Landau parameter but not on the special material.

 T_c , but it can be assumed that this mechanism is effective in the whole temperature range. This is not the case for the attraction caused by field reversal because nonlocal effects are important only at low temperatures. As the transition temperature is approached the penetration depth $\lambda_L \sim (1 - t)^{-1/2}$ (which is a characteristic length for magnetic field variations) becomes very large and all materials behave like London superconductors.

Summarizing, one can say that the existence of type-II superconductors with a first-order phase transition at H_{c1} is well established experimentally. Present theoretical calculations, however, give only a qualitatively correct picture of the observed

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