Influence of Impurities on the Laser Induced Breakdown in Liquid He^4

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Dedicated to Professor Dr. H. Maier-Leibnitz on his 60th birthday

The threshold power for laser induced breakdown in liquid He^4 has been investigated above and below the λ-transition. The results seem to indicate that the threshold power is mainly determined by the impurity content of the liquid.

Recent experiments [1–4] on laser induced breakdown in compressed helium gas and in liquid helium suggest that optical breakdown is caused by an electronic avalanche mechanism [5]. According to this mechanism the threshold power for optical breakdown should increase with decreasing electronic scattering rate, or with atomic density since electrons are mainly scattered by atoms. This close relationship between the threshold power and the atomic density has indeed been observed both in helium gas [1–3] and in liquid helium from 4.2 °K to 2.2 °K [3, 4].

At lower temperatures, however, WINTERLING, HEINICKE and DRANSFELD [4] observed a strong temperature variation of the threshold power, which increased by one order of magnitude on cooling from T_λ to 1.5 °K, although the atomic density remains almost constant in this temperature interval.

In this communication we present evidence that this anomalous temperature dependence is not caused by a temperature dependent electronic scattering rate as suggested previously [4, 6] but by the impurity content of the liquid.

The Role of Initial Electrons

ABRIKOSOVA and BOCHKOVA [3] as well as WINTERLING and HEINICKE [7] have observed an interesting time dependence of the threshold power: The initial optical breakdown required the highest available laser power, while subsequent breakdowns a few minutes later could be produced by a power which was about one order of magnitude smaller. This reduction only disappeared if the time interval between pulses was larger than 30 minutes.

Theoretically, optical breakdown in pure helium can only occur at moderate threshold powers *if there is at least one free electron* in the focal volume [5] of the liquid when the laser pulse is arriving. Therefore the question arises whether the relatively high threshold power observed for the first breakdown is simply caused by the lack of an initial electron, and whether the electrons — left over from the first breakdown — are responsible for igniting the subsequent breakdown at a much lower threshold power.

In order to check this assumption we injected electrons from a hot cathode [8] into the liquid sweeping them into the focal volume by an electric field (50 V/cm). From the measured current density ($2 \cdot 10^{-11}$ A/cm^2) and the known electronic mobility [9] the number of electrons in the focal volume was estimated to be at least 10^3. This high density of electrons (6 orders of magnitude higher than the natural background [10]) produced no observable influence on the threshold power neither for the first nor for subsequent pulses. — WINTERLING, HEINICKE and DRANSFELD [4] reached essentially the same conclusion from their experiments: By a dc electric field they swept away all charge carriers out of the focal volume. This removal of electrons prior to inducing breakdown had again no influence on the observed threshold power.

The Role of Impurities

From the above observations one can safely conclude that a considerably higher density of free electrons must be necessary for the production of breakdown. Presumably impurities alone, having a much lower ionisation energy than helium atoms, produce the first copious supply of electrons when the laser pulse arrives.

If no special precautions are taken to purify liquid helium, it may have an impurity content of one part per million or more, not only of He^3 but also of microcrystallites of air and other materials which can be easily ionized in the initial phases of each laser pulse. If the number of free carriers generated this way inside the focal volume exceeds about 10^6 the threshold power for optical breakdown is expected to decrease noticeably [5].

Since these foreign materials have mostly a density higher than that of the liquid, they are able to sediment out, only rather slowly in He I, but relatively fast in superfluid He II [11].

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If the breakdown is indeed promoted by sedimenting impurities one would except the threshold power to increase in time if the liquid is kept in the superfluid state:

1) When the liquid was maintained at 1.9 °K for 30 minutes we observed an increase of the threshold power by one order of magnitude.
2) At the same temperature the threshold power decreased again by similar amount if the sedimentation of the impurities was prevented, for example, by simply stirring the liquid mechanically.

All these observations lead to the conclusion that the presence of impurities in liquid helium plays a very important role in the observed process of optical breakdown.

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Curie-Weiß Temperature Dependence of Paraelasticity *

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Anelastic strains exceeding elastic strains up to a factor of 30 have been observed close to the critical point of H in Nb. The temperature dependence of the relaxation strength follows a Curie-Weiß law for temperatures

$$(T-T_c)/T_c \geqq 6\cdot 10^{-3}\,.$$

I. Introduction

The Curie-Weiß type temperature dependence of magnetic or dielectric susceptibilities as well as of the compressibility of real gases have been known for long time. Only recently the analogous temperature dependence has been discovered for the elastic response of a solid, namely niobium loaded with hydrogen [1]. Actually this system shows elastic properties which are a mixture of those of a solid and a real gas. This peculiar behaviour results from the fact that hydrogen in metals like Nb behaves like a gas in the space of the metallic host lattice [2]. The phase diagram for H in Nb [3] shows a miscibility gap separating the lattice-gas- and lattice-liquid-phases and ending in a critical point.

The properties of the lattice gas can be studied experimentally by means of the elastic diffusional relaxation process (Gorsky effect) [4]. The relaxation strength Δ_E of the Gorsky effect is given by [5]

$$\Delta_E = \frac{\varepsilon_a}{\varepsilon_e} \sim \left(\frac{\partial \mu}{\partial \varrho}\right)^{-1}_{iso} \sim \varkappa_{iso}\,,$$

ε_e = elastic strain, ε_a = total anelastic strain, μ = chemical potential, ϱ = density of the lattice gas, $\varkappa_{iso}$ = isothermal compressibility of the lattice gas.

For the critical density ϱ_c the compressibility approaches infinity for $T \to T_c$. Therefore the relaxation strength should diverge as it approaches the critical point.

II. Experiments

Static Gorsky-effect measurements have been performed in a torsion pendulum apparatus. The sample consisted of a Nb-spring, loaded with the critical concentration of 34 at.-% H. Both the relaxation strength and the relaxation time have been measured as a function of temperature. More experimental details have been published in Ref. [6].

1) Relaxation Strength

Usually, elastic after-effect phenomena are at maximum in the percent region of the elastic strain. Fig. 1 shows as a counter-example an elastic after-effect curve as measured for a $NbH_{0.34}$ — sample at 181 °C. The anelastic strain ε_a exceeds the elastic strain by a factor of 20, and still remains fully reversible. Fig. 2, in which

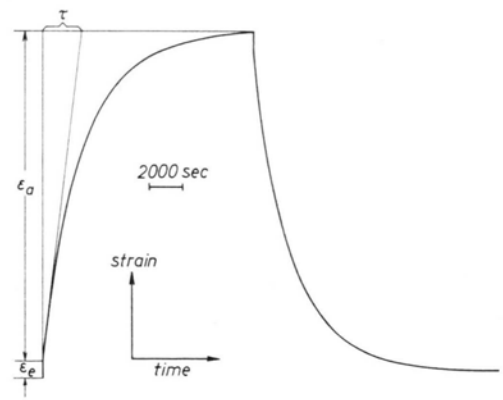

Fig. 1. Recorder chart of a measured elastic after-effect curve for Nb+34 at.-% H at 181 °C.

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* Troubles caused by elastic after-effects of gold foils in a gold-leaf electrometer were the starting point for a research program, initiated by Prof. H. MAIER-LEIBNITZ, finally leading to the discovery of ferro-elasticity.

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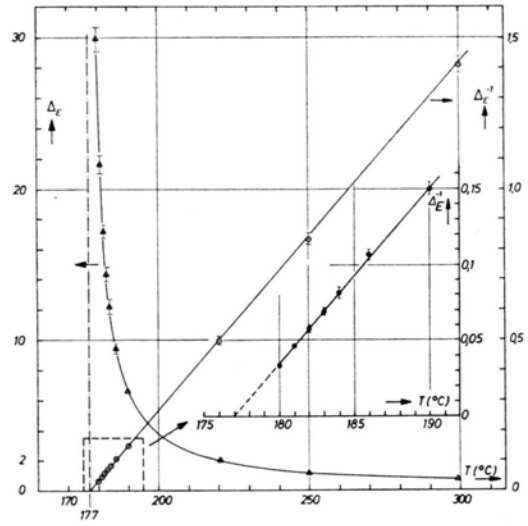

Fig. 2. Relaxation strength and reciprocal relaxation strength as a function of temperature.

the relaxation strength Δ_E is plotted versus temperature, shows the divergence of ε_a as it approaches T_c. To avoid nonlinear anelastic effects the total strain was always kept in the region of 10^{-5} by reducing the applied stress corresponding to the increase in Δ_E, thus keeping the gradient in hydrogen concentration approximately constant. The plot Δ_E^{-1} versus temperature (Fig. 2) yields a Curie-Weiß law for the temperature range investigated. By an extrapolation to $\Delta_E^{-1}=0$ one obtains a critical temperature $T_c=(177\pm0.5)\,^\circ$C.

2) The Diffusion Coefficient

According to Eq. (51) in Ref. [5] the diffusion coefficient D is given by

$$D = B\,\varrho\,(\partial\mu/\partial\varrho)$$

[$B(\varrho, T)$ = mobility]. Usually the mobility determines the temperature dependence of D. Close to a critical point the temperature dependence of $\partial\mu/\partial\varrho$ becomes significant resulting in "critical slowing down". Using the relation 13.55 $D\tau = d^2$ (d = diameter of cylindrical sample) the diffusion coefficient D can be determined from the relaxation time τ. Fig. 3 shows the strong decrease of D near T_c. The largest experimental relaxation time was about 3000 sec, this limit being given by electronic and temperature drifts.

In the upper curve of Fig. 3 the diffusion coefficient D is multiplied by $T/(T-T_c)$. By this procedure the singular part of the temperature dependence resulting from $\partial\mu/\partial\varrho$ is eliminated. The thus corrected diffusion coefficient, representing essentially the temperature dependence of the mobility B, shows no singularity on approach of the critical point. It is interesting to

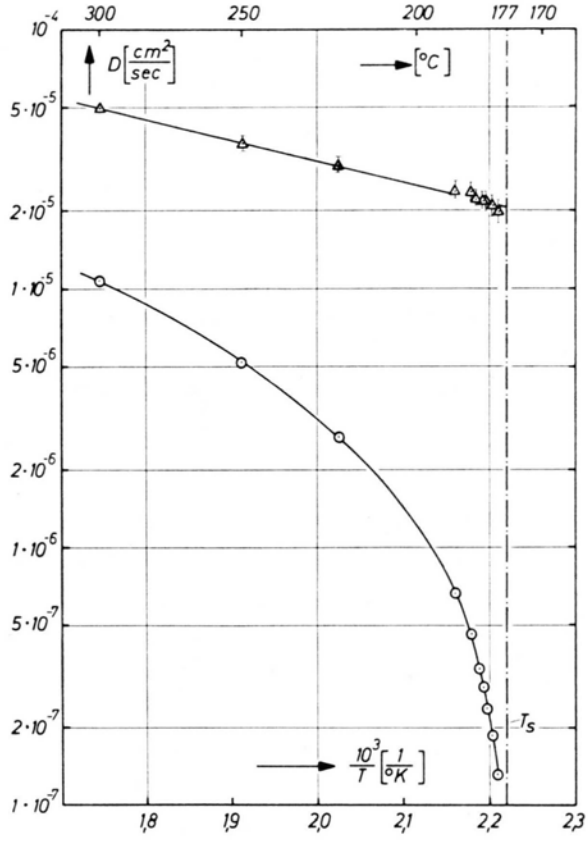

Fig. 3. Logarithm of the diffusion coefficient as a function of the reciprocal temperature (lower curve). For upper curve see explanation in the text.

note that the absolute value at 300 °C coincides with that measured for infinite dilution [6], whereas the temperature dependence is appreciably stronger [$U=(0.15\pm0.01)$ eV, $D_0=(3\pm2)\cdot10^{-3}$ cm²/sec compared to $U=(0.106\pm0.006)$ eV, $D_0=(5\pm1)\cdot10^{-4}$ cm²/sec for infinite dilution].

III. Discussion

The essential results of this paper are the following:

1) No deviation from the Curie-Weiß type temperature dependence for $(T-T_c)/T_c \geqq 6\cdot10^{-3}$ (as demonstrated in the enlarged insert in Fig. 2).

2) No singularity in the mobility B for the same temperature range.

Two reasons can be stated which make the validity of the mean field approximation this close to the critical point plausible:

a) There exists strong evidence that the interaction responsible for the condensation of the lattice gas is

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the elastic dipole-dipole interaction [7, 8] which is long range, thus favoring the mean field approximation [9].

b) The essential shortcoming of Landau's theory is the neglect of the interaction of fluctuations. On the other hand there are reasons to assume that in the system studied critical fluctuations with wavelength short compared to sample size are suppressed due to coherency strains in the host lattice [10]. Therefore, if only very few modes of the density fluctuations become critical, the interaction of the fluctuations may indeed be neglected.

The nonsingular behaviour of the mobility may also be attributed to the nonsingular behaviour of nearly all of the fluctuation modes.

Acknowledgments

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Die Emission geladener Teilchen nach dem Einfang langsamer Neutronen in ^{22}Na

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Der Einfangsquerschnitt für thermische Neutronen von ^{22}Na, einem Positronenemitter mit $T_{1/2} = 2{,}6$ a beträgt 35.9 ± 1.2 kb [1, 2]. Im gegenwärtigen Experiment wurde das Spektrum der geladenen Teilchen (s. Abb. 1) nach dem Einfang langsamer Neutronen untersucht.

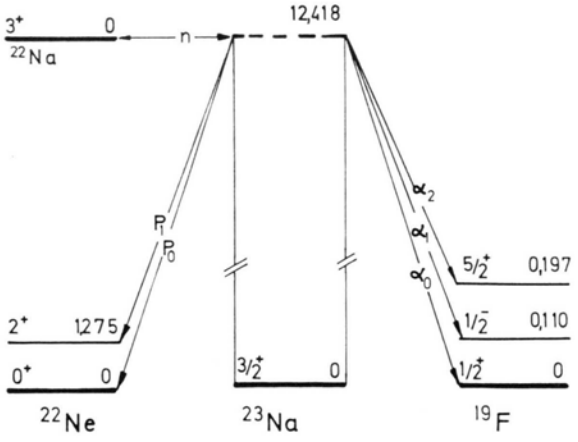

Abb. 1. Reaktionen nach dem Einfang langsamer Neutronen in ^{22}Na.

Die Messungen wurden durchgeführt am Strahlrohr QRII des Forschungsreaktors München bei einem Fluß von 3×10^6 n/sec·cm^2. Der mittels ^{6}LiF-Absorbern auf einen Durchmesser von 0.5 cm abgeblendete Strahl fiel auf eine ^{22}Na-Quelle der Aktivität 2 μCi/cm^2, die durch Bedampfen einer Aluminiumfolie mit trägerfreier $^{22}Na_2CO_3$-Lösung hergestellt wurde. Die geladenen Teilchen wurden mit einem Si-Detektor (100 mm^2 Fläche, max. 300 μm Dicke) nachgewiesen, der in einer Vakuumkammer außerhalb des Neutronenstrahls aufgestellt war (s. Abb. 2). Zur Verringerung der Untergrundzählrate, verursacht durch Target und Strahlrohr, wurde die Spannung am Detektor entsprechend dem Nachweis einer maximalen Protonenenergie von 4 MeV gewählt, also größer als $E(p_0)$.

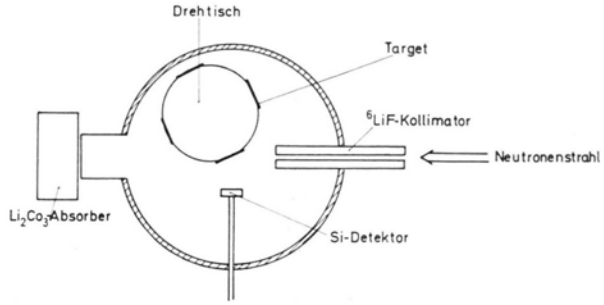

Abb. 2. Schematische Ansicht der experimentellen Anordnung.

Abb. 3 zeigt ein Spektrum der geladenen Teilchen aus der (^{22}Na + n)-Reaktion. Während die Protonengruppe von 2.25 MeV vom Zerfall des Compoundkerns ^{23}Na zum ersten angeregten Zustand von ^{22}Ne als sehr starke Linie im Spektrum auftritt, konnten weder die Protonengruppe zum Grundzustand von ^{22}Ne noch die bei der Reaktion ^{22}Na(n, α)^{19}F entstehenden α-Teilchen beobachtet werden. Die beiden Linien bei den Energien 1.43 MeV bzw. 1.77 MeV wurden als α-Gruppen der

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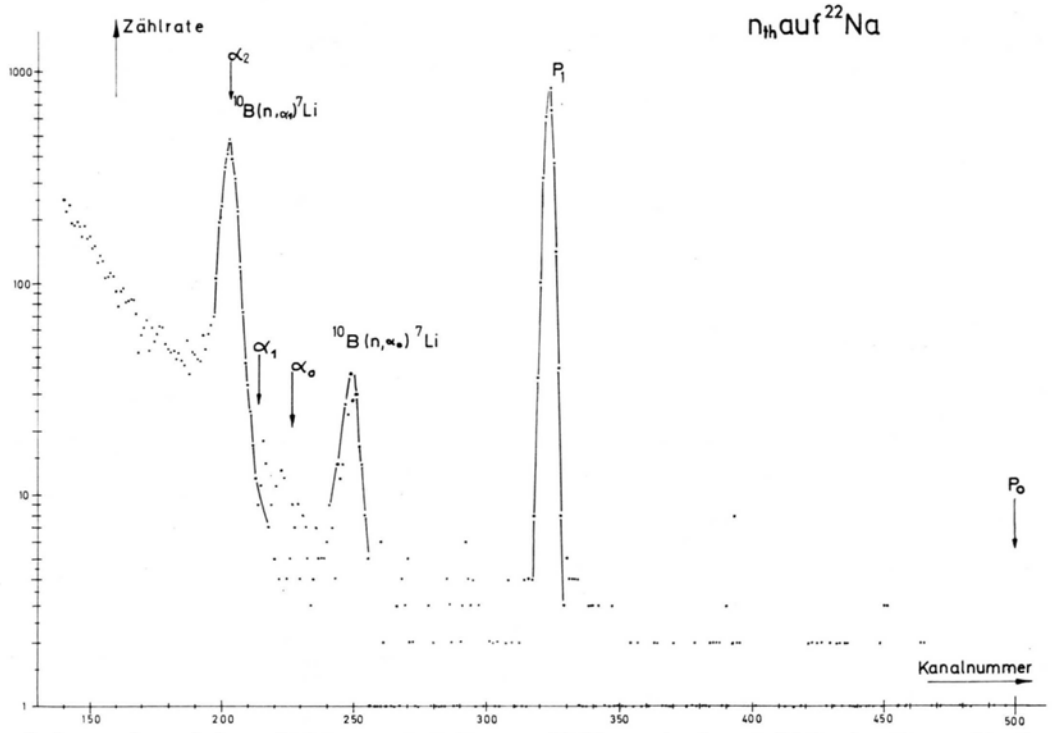

Abb. 3. Spektrum der geladenen Teilchen nach 7 Tagen Meßdauer. An den mit Pfeilen bezeichneten Kanälen werden α_0, α_1, α_2 bzw. p_0 erwartet.

unerwünschten Reaktion $^{10}B(n,\alpha)^7Li$ identifiziert. Das System wurde geeicht mit α-Teilchen aus dem Zerfall von ^{242}Cm bzw. ^{238}Pm, sowie mit den bei der Reaktion $^6Li(n,\alpha)^3H$ entstehenden Partikeln. Aus unseren Daten ergibt sich der (n,p_1)-Wirkungsquerschnitt zu $(4\pm2)\times10^4$ b.

Der große thermische Neutroneneinfangsquerschnitt von ^{22}Na kann auf die Existenz eines Niveaus nahe der Neutronenschwelle von ^{23}Na zurückgeführt werden. Mit großer Wahrscheinlichkeit ist dies eine bei der Reaktion $^{19}F(\alpha,p)^{22}Ne$ bei 4 ± 4 keV unterhalb der Neutronenschwelle gefundene Resonanz[3], welche ebenso wie der Neutroneneinfangszustand in diesem Experiment, stark durch Protonenemission zum ersten angeregten Zustand von ^{22}Ne zerfällt. Spin und Partität dieser Neutronenresonanz können dabei $5/2^+$ oder $7/2^+$ sein. Bei der $^{19}F(\alpha,p)^{22}Ne$-Reaktion ergab sich für die Breite des α_0-Kanals $[230/(2J+1)]\times(\Gamma_T/\Gamma_{p1})$ eV (Γ_T bzw. Γ_{p1} sind die totale Breite bzw. die Breite des p_1-Kanals). Dieser Wert ist größer als das WIGNER-Limit[4] der α_0-Breite für $J^\pi=7/2^+$ von 3 eV: Spin und Parität sind also wahrscheinlich $5/2^+$.

Ein weiterer Vergleich unserer Ergebnisse mit den Resultaten des (α,p)-Experiments zeigt jedoch eine Diskrepanz: die Intensität des α_0-Übergangs im vorliegenden Versuch kann mit Hilfe der (α,p)-Daten mittels einer Breit-Wigner-Formel für eine isolierte Resonanz abgeschätzt werden:

$$\Gamma_{\alpha0}/\Gamma_{p1}=(230/6\,\Gamma_T)\times(\Gamma_T/\Gamma_{p1})^2.$$

Da die totale Breite aufgrund der Resultate des (α,p)-Experiments kleiner als 6 keV ist, sollte das Verhältnis der Intensitäten des α_0-Übergangs zum p_1-Übergang größer sein als $6{,}2\times10^{-3}$. Unsere Ergebnisse lassen dagegen auf ein Verhältnis von $(1\pm1{,}5)\times10^{-3}$ schließen.

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