Inelastic electron lifetime in niobium films

Bruce J. Dalrymple, Stuart A. Wolf, Alexander C. Ehrlich, and Darrel J. Gillespie
Naval Research Laboratory, Washington, D.C. 20375-5000
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The perpendicular magnetoconductance has been measured for thin niobium films ($R_\square = 47$–218 Ω) for temperatures from near the superconducting transition temperature T_c to 20 K. The electron phase-coherence lifetime has been determined by analyzing magnetoconductance data, using the theories of localization and superconducting fluctuation effects in thin films. This electron lifetime is interpreted in terms of electron-electron and electron-phonon scattering mechanisms.

I. INTRODUCTION

Recently the transport properties of disordered, thin (quasi-two-dimensional) metallic films have attracted a great deal of theoretical and experimental attention.[1] At low temperatures, quantum corrections to the conductivity become important. Localization of the electron wave functions and changes in the character of the electronic screening produce effects not predicted by the standard, quasiclassical transport theory. Studies of the magnetoconductance (MC) in the weakly localized regime are of particular interest. The theory here is reasonably well developed and analysis of MC data can yield the electron phase-coherence lifetime τ_ϕ. However, although many studies of thin films of both superconducting and nonsuperconducting metals have been reported, the identification of the specific microscopic mechanisms contributing to τ_ϕ remains an open question in many cases.

In this paper we report measurements of the superconducting transition temperature T_c and perpendicular MC (for $T > T_c$) made on a series of high quality, thin, sputtered niobium films. The MC of these films is in good agreement with the theory of localization and superconducting fluctuations in thin films, and has been analyzed to determine τ_ϕ. The temperature dependence of τ_ϕ can be explained by invoking a combination of electron-electron and dirty-limit, two-dimensional electron-phonon scattering processes. In addition, the magnitude of τ_ϕ is in reasonable quantitative agreement with this model for thicker films (thicknesses > 50 Å). However, we observe a systematic discrepancy between the calculated and experimental magnitudes of τ_ϕ for thinner films.

II. MAGNETOCONDUCTANCE THEORY

The magnetoconductance theory relevant for our samples is a combination of weak localization theory and Maki-Thompson and Aslamasov-Larkin superconducting fluctuation theories. The contributions to the MC caused by two-dimensional Coulomb interaction effects[2] are negligible here. Zeeman splitting effects[3] are also negligible, in part because of the small electron spin-orbit scattering lifetime τ_{SO} (see below). The expression for the two-dimensional localization contribution to the magneto-conductance per square is[4,5]

$$\Delta G_{\rm loc}(H) = \frac{e^2}{2\pi^2\hbar}[1.5F(H_2/H) - 0.5F(H_3/H)] \ . \quad (1)$$

Here, $F(X) = \Psi(0.5+X) - \ln(X)$, where Ψ is the digamma function. $H_2 = \frac{4}{3}H_{SO} + H_\phi + \frac{2}{3}H_s$ and $H_3 = H_\phi + 2H_s$. The normalized scattering rates H_ϕ, H_{SO}, and H_s are given (in gauss) by $H_x = \hbar c/(4eD\tau_x)$, where τ_x is the corresponding electron lifetime and D is the diffusion constant. τ_s is the lifetime due to scattering from paramagnetic impurities. The Maki-Thompson (MT) MC is proportional to the localization term when the spin-orbit contribution is removed:[5,6]

$$\Delta G_{\rm MT}(H) = -\beta(T,T_c)\Delta G_{\rm loc}(H;\tau_{SO}=\infty) \ . \quad (2)$$

$\beta(T,T_c)$ is a function introduced by Larkin.[6] The use of this field-independent $\beta(T,T_c)$ is restricted to low fields,[7]

$$H < H_{\rm lim} \equiv (k_B cT/4De)\ln(T/T_c) \ .$$

The Aslamasov-Larkin (AL) MC is given by[8]

$$\Delta G_{\rm AL}(H) = \frac{e^2}{16\hbar\epsilon}\{2X^2[\Psi(1/2+X/2) - \Psi(1+X/2) + 1/X] - 1\} \ . \quad (3)$$

Here, ϵ is the reduced temperature which we define, following Bergmann,[9] to be $\epsilon = \ln(T/T_c)$. The quantity X is defined by $X = \epsilon/h$, where h is the reduced field $h = (2e/\hbar c)\xi(0)^2 H = 9.044\times10^{-5}DH/T_c$. The last form of this relationship is obtained using the standard BCS theory results: $\epsilon(0)^2 = 0.72\xi_0 l$ and $\xi_0 = \hbar v_F/(1.764\pi k_B T_c)$.

The total theoretical formula for the MC per square is $\Delta G(H) = \Delta G_{\rm loc}(H) + \Delta G_{\rm MT}(H) + \Delta G_{\rm AL}(H)$. These formulas are defined such that $\Delta G(0) = 0$.

III. SAMPLE PREPARATION

The niobium films were produced by rf sputtering in an ion-pumped vacuum system with a base pressure of 5×10^{-9} torr. Both quartz and sapphire substrates have been used. A deposition rate of 90 Å/min was obtained using 900 W of rf power and a purified-argon pressure of 6×10^{-2} torr. During film deposition the ion pump is valved off, but the deposition chamber is still pumped by

a Ti sublimation pump and liquid-nitrogen-cooled cryopanels. Taking T_c for a given thickness as an indicator of sample quality, the best films (the highest T_c's) were produced on sapphire substrates using the following procedure: During the initial pumpdown of the system, the substrates are baked at ~800 °C using carbon-strip heaters located below the molybdenum substrate table. Just prior to deposition the substrates are baked at ~450 °C for 15 min to effect a final cleaning of the surfaces. The substrate temperature during deposition is ~200 °C.

A 100-Å niobium film sputtered onto an amorphous carbon substrate was examined by electron diffraction.[10] This showed the film to have the bcc crystal structure with the bulk lattice constant $a = 3.30 \pm 0.01$ Å. The diffraction results, along with dark field electron microscopy results, imply a uniform distribution of grain orientations, with no pronounced texturing.

The films are patterned using standard photolithographic techniques to produce strips 1 mm wide, with contacts for making four-terminal resistance measurements. The excess metal is removed using an argon-ion mill, with a photoresist pattern serving as the etch mask. The resulting sample geometry has 3.5 squares between the voltage leads. The nominal film thickness is determined by anodization in a room-temperature, saturated boric acid solution. The voltage across the oxide produced by the anodization process is related to the thickness of the metal removed by the factor 6.6 Å/volt.[11] One film produced in each deposition run is anodized completely to determine the thickness of the batch. As expected, the thickness determined in this manner correlates very well with the sputtering time.

The anodization process has also been used to reduce the thickness of several of the films used in this study. Our data for T_c and for the MC, and the parameters obtained by analyzing these data indicate no difference between the anodized and unanodized films. We therefore conclude that the oxide layer produced by the anodization process is inert with respect to the electronic properties relevant to our experiments. The unanodized films also acquire a thin oxide surface layer (approximately 5 Å thick) during processing.

IV. EXPERIMENTAL RESULTS

Measurements of the resistance as a function of temperature and magnetic field have been carried out in a dewar equipped with a 95-kG superconducting magnet. The dc measuring current was kept low enough to avoid sample heating effects.

Figure 1 shows T_c as a function of the resistance per square $R_\square$ for films with thicknesses in the range 22 to 135 Å. T_c is defined by the midpoint of the resistive transition. The transition widths (10—90%) are typically 0.2 K. $R_\square$ is defined at a temperature several degrees above T_c, where the enhancement of the conductivity due to superconducting fluctuations is negligible and $R(T)$ is almost constant. T_c decreases linearly as $R_\square$ increases, at a rate of 37 mK/Ω.[12] Previous studies of superconductivity in thin niobium films have emphasized the dependence of T_c on thickness.[11,13] Considered as a function of thickness, the T_c's of the films shown in Fig. 1 are in good agreement with these earlier studies.

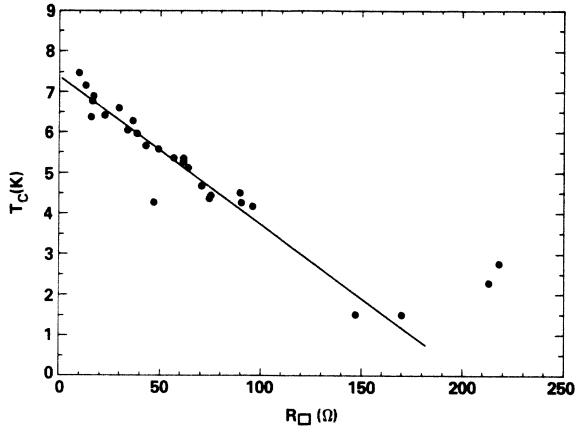

FIG. 1. T_c as a function of $R_\square$ for Nb films with nominal thicknesses in the range 22 to 135 Å. The solid line is a least-squares fit to the data with $R_\square < 200$ Ω and has a slope of -0.037 K/Ω. The two films with $R_\square > 200$ Ω are nominally 22 Å thick. That these two points do not fall on the same line with the other data may be a consequence of the nonuniform morphology of the thinnest samples.

Above T_c, $R(T)$ is a monotonically increasing function of temperature due to the dominating influence of the superconducting fluctuations. The residual resistance ratio of the films varies from 1.1 to 1.6, being higher for films with lower $R_\square$.

At this time, no completely satisfactory explanation exists for the suppression of T_c in thin-film superconductors. Theories based on the proximity effect[11,14] and also on electron interaction effects[15] have been proposed. Most of the data existing in the literature are for amorphous alloys or quenched condensed elements.[16] In these highly disordered systems the resistivity is typically several hundred $\mu\Omega$ cm and the rate of decrease of T_c with increasing $R_\square$ is quite uniformly in the range 4—7 mK/Ω. Also, these data are frequently obtained, at least in part, on rather thick films where the dimensionality of the observed effects is not clear. In contrast, our Nb films have much lower resistivities, 20—50 $\mu\Omega$ cm, and are thinner ($d < 125$ Å), so that the two-dimensional (2D) limit more clearly applies. These differences, especially the higher crystalline quality of our films, may explain the fact that the rate of decrease of T_c with increasing $R_\square$, 37 mK/Ω, is considerably larger than has been reported for more disordered systems.

The MC of seven films have been studied in detail. The properties of these samples are listed in Table I. Figure 2 shows MC data at several temperatures for a typical sample, along with the theoretical fits. The MC is negative at all temperatures and at all accessible magnetic fields, as expected for a superconductor with a short τ_{SO}. For all the films studied, the thickness d is considerably less than both the inelastic diffusion length $L_T = (D\tau_\phi)^{1/2}$ and the magnetic field length scale[17,18] $L_H = (\hbar c/4eH)^{1/2}$ (at the values of H used to determine τ_ϕ). Therefore, the two-

TABLE I. Parameters of the samples for the magnetoconductance measurements, listed in order of decreasing nominal thickness. Samples $S1$, $S2$, and $S4$ were deposited on quartz substrates; all the other samples are on sapphire substrates.

Sample	$R_\square$ (Ω)	d (Å)	T_c (K)	D (cm^2/sec)	τ_s (psec)	$\frac{B_{calc}}{B_{expt}}$
$S1$	47	70	4.26	0.9	88	1.49
$S2$	71	55	4.67	0.9	53	1.55
$S3$	75	39	4.42	1.0	84	2.91
$S4$[a]	62	32	5.35	1.2	77	3.39
$S5$	170	26	1.50	1.2	94	4.15
$S6$	218	22	2.75	1.0	53	4.07
$S7$[a]	90	22	4.49	1.3	37	4.36

[a]Sample formed by thinning a thicker sample by anodization. Original thicknesses: $S4=135$ Å and $S7=55$ Å.

dimensional localization formalism should be applicable. The diffusion constant D has been obtained from the slope of the perpendicular critical field using the standard dirty-limit theory.[19]

In previously published studies, the effect of Aslamasov-Larkin fluctuations has not usually been included in the analysis of the MC of thin films. However, we find that this contribution is not completely negligible. Leaving it out results in an approximate 5% increase in the inferred values of τ_ϕ with, however, little change in the temperature dependence.

The MC theory depends on the parameters T, T_c, and D, which are known, and on τ_{SO}, τ_s, and τ_ϕ, which are to be determined by fitting the theoretical expression for $\Delta G(H)$ to the data. However, the best fits are always obtained for $\tau_{SO} \ll \tau_\phi$. In this limit the theory is insensitive to the exact value of τ_{SO}, so this parameter cannot be determined with any accuracy.[20] Therefore, the theory is fit to the MC data with τ_ϕ and τ_s as the only adjustable parameters. τ_s is assumed to be temperature independent, while τ_ϕ is allowed to vary as a function of temperature. In obtaining the fits, only data that fall below a cutoff field are used. This cutoff field, which is always less than the limit on the Larkin theory H_{lim}, is chosen to be as large as possible, consistent with obtaining good agreement at all the experimental points included. This cutoff field is typically chosen to be approximately $\frac{1}{2}H_{lim}$. However, the inferred values of τ_ϕ are not sensitive to the exact value of the cutoff field. With this procedure, reliable values of τ_ϕ can be obtained down to about 1 K above T_c.

V. ANALYSIS OF THE ELECTRON PHASE-COHERENCE LIFETIME

Figure 3 shows the phase-coherence scattering rate τ_ϕ^{-1} as a function of temperature for a typical sample. These data cannot be fit to a simple power law of the form $\tau_\phi^{-1} \propto T^n$ for any value of n. The need for including at least two powers of T is obvious if $\tau_\phi(T)$ is examined on a

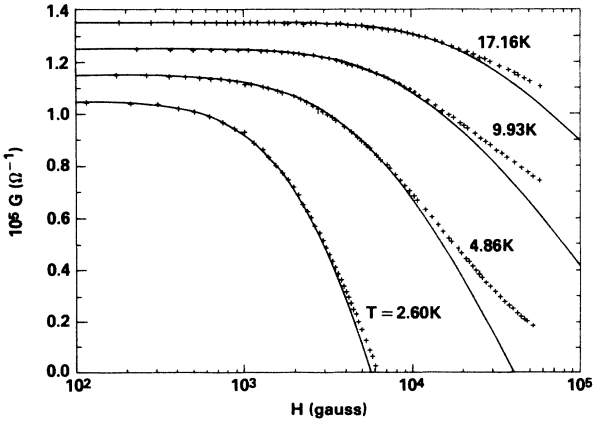

FIG. 2. Magnetoconductance of sample $S5$ at several temperatures. The calculated MC is also shown. For clarity the data for each temperature are offset by an arbitrary amount so that the curves do not intersect. Also, only a fraction of the experimental points is shown at each temperature.

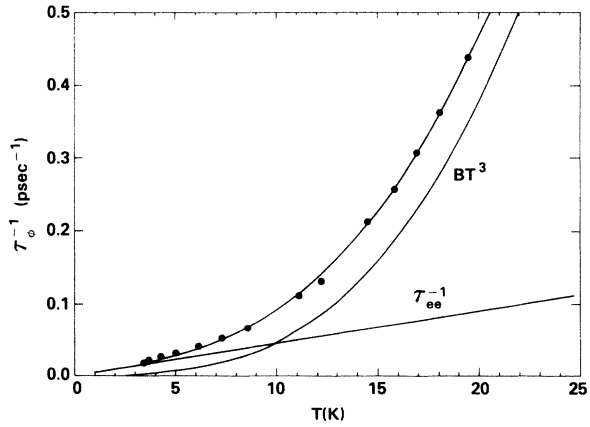

FIG. 3. Electron phase-coherence scattering rate as a function of temperature for sample $S6$. The total theoretical fit is shown along with its two components: the scattering rate due to electron-electron processes and the term BT^3 ascribed to the electron-phonon interaction.

log-log plot, as the data clearly display different slopes at high and low temperatures. The log-log plot indicates that τ_ϕ^{-1} is a linear function of T at the lowest temperatures, with a crossover to a higher power of T at higher temperatures. It now appears well established that there is a contribution to τ_ϕ^{-1} due to electron-electron interactions which is linear in T.[1,5,21] We have therefore fit the experimental data to a function of the form $\tau_\phi^{-1}(T)=\tau_{e\text{-}e}^{-1}(T)+BT^n$, as shown in Fig. 3. Here, B and n are parameters to be determined, and $\tau_{e\text{-}e}^{-1}(T)$ is the phase-coherence lifetime due to electron-electron interactions,[21] which is linearly proportional to T,

$$\tau_{e\text{-}e}^{-1}=\frac{e^2R_\square}{2\pi^2\hbar^2}k_BT\ln\left[\frac{\pi\hbar}{e^2R_\square}\right]. \tag{4}$$

An exponent of $n=3$ provides a better fit to the data than any other integer.[22] This BT^3 term in τ_ϕ^{-1} can be most naturally ascribed to electron-phonon scattering. A cubic temperature dependence is expected from the theory of inelastic electron-photon scattering in the dirty, two-dimensional limit where $\tau_{e\text{-}p}^{-1}$ is given by[23]

$$\tau_{e\text{-}p}^{-1}=14\pi^2\zeta(2)\frac{k_B\lambda_{e\text{-}p}l}{\hbar d\Theta_D}T^3. \tag{5}$$

Here $\lambda_{e\text{-}p}$ is the electron-phonon coupling constant, Θ_D is the Debye temperature, and ζ is the Riemann ζ function. For Nb we take[24] $\lambda_{e\text{-}p}=1$ and[25] $\Theta_D=277$ K. The elastic mean free path l is obtained from $D=v_Fl/3$ with[26] $v_F=2.7\times10^7$ cm/sec. The mean free path determined in this manner varies only slightly; from 10 to 14 Å.

The phonon dimensionality is set by the most probable phonon wave vector $q_T=k_BT/(\hbar v_s)$, where v_s is the speed of sound.[4,5,23] Given the nonspherical nature of the Nb Fermi surface, the transverse speed of sound[27] $v_s=2.2\times10^5$ cm/sec may be the appropriate value to use here. In this case, the phonon wavelength λ_T, corresponding to q_T, varies from 263 Å at 4 K to 53 Å at 20 K. For most of the samples $\lambda_T>d$ at all temperatures. However, for samples $S1$ and $S2$, $\lambda_T<d$ at the highest temperatures of interest. (The wavelength corresponding to the longitudinal speed of sound[27] $v_s=5.1\times10^5$ cm/sec is considerably larger than d for all samples at all temperatures.) The question of phonon dimensionality is complicated by the coupling of the acoustic-phonon modes to the substrate. However, this coupling appears to be fairly weak: The transverse-phonon transmission coefficients for Nb to quartz and to sapphire are[28] 0.21 and 0.091, respectively. Therefore, the two-dimensional limit appears at least marginally justified. The dirty limit unambiguously applies since $\lambda_T\gg l$ for all films.

The coefficient of T^3 calculated using Eq. (5) is compared with the experimental values in Fig. 4, where both are shown as a function of thickness. The ratio of calculated to experimental coefficients $B_{\text{calc}}/B_{\text{expt}}$ is included in Table I. For the thicker films with $d>50$ Å, the calculated and measured electron-phonon scattering rates are in fair agreement. However, there is clearly a systematic discrepancy for the thinner films. The coefficient

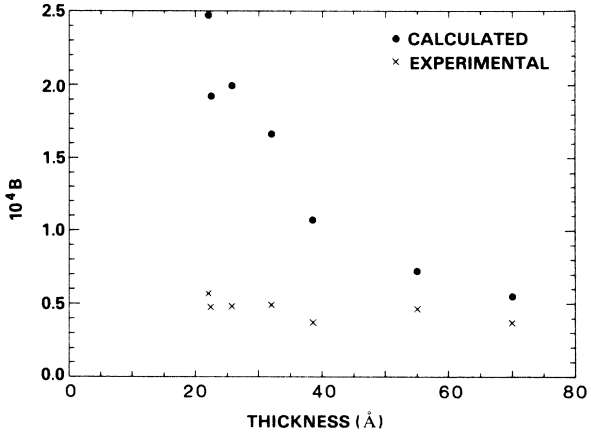

FIG. 4. Calculated and experimental coefficients of the T^3 term in $\tau_\phi^{-1}(T)$, as a function of sample thickness.

calculated using Eq. (5) is proportional to $1/d$, while the experimental coefficient increases very slowly, if at all, as the thickness decreases. This may be in part related to the film morphology. The grain size in the films we have studied is[29] approximately 40–50 Å, so films with thicknesses of this order or less can be expected to have pronounced local thickness variations. The values used above for d are determined by anodization and sputtering time and are therefore characteristic of the average sample thickness. The use of this average thickness provides semiquantitative agreement with the experimental results only for the relatively thicker films.

Values for the magnetic scattering lifetime τ_s are shown in Table I. If spin scattering is neglected, the inferred temperature dependence of τ_ϕ^{-1} becomes less than linear in T at the lowest temperatures. However, there is no known temperature-dependent scattering mechanism which could cause such a weak temperature dependence. Several studies have been published where values of τ_s have been correlated with the concentration of magnetic impurities.[30] Based on these results, we can estimate that our films contain on the order of 10 ppm magnetic impurities. Magnetic impurities at this concentration level may exist in the Nb sputtering target or may conceivably be introduced during film deposition.[31]

The lowest-temperature τ_ϕ data are obtained close to T_c. Recently, Brenig, Chang, Abrahams, and Wölfle[32] have shown that electron scattering involving superconducting fluctuations can contribute to τ_ϕ^{-1}, producing an upturn near T_c. To lowest order, this mechanism could have the same effect as magnetic impurities. However, this scattering involving superconducting fluctuations is sensitive to a magnetic field, and should be largely suppressed at the fields at which the relevant structure occurs in the MC of our samples. Further evidence that scattering from superconducting fluctuations is not present is provided by data for Nb films which do not superconduct because they contain gaseous impurities. For these films $\tau_\phi(T)$ can also be fit to the formula[33] $\tau_\phi^{-1}(T)=\tau_{e\text{-}e}^{-1}(T)+BT^3$, with comparable values of τ_s.

VI. CONCLUSIONS AND COMMENTS ON PREVIOUS WORK

We have shown that the MC of thin, superconducting Nb films is in good agreement with the theory of localization and superconducting fluctuation effects. The electron phase-coherence lifetime has been determined for seven films covering a range of $R_\square$'s and thicknesses. For the thicker films, $\tau_\phi^{-1}(T)$ agrees to within a factor of 2 with a theoretical model consisting of two terms: The electron-electron phase-coherence lifetime, given by Eq. (4), combined with a term due to electron-phonon interactions, which we have assumed is given by the formula for inelastic electron-phonon scattering in the dirty, two-dimensional limit.

For thinner films, $d < 50$ Å, the theoretical model for $\tau_\phi^{-1}(T)$ does not work as well. The scattering rate $\tau_\phi^{-1}(T)$ can still be fit to the equation $\tau_\phi^{-1}(T)=\tau_{e\text{-}e}^{-1}(T)+BT^3$, but the magnitude of the coefficient B does not increase as the thickness decreases in the manner predicted by Eq. (5). It seems most probable that this discrepancy is caused by the inadequacy of the electron-phonon scattering theory we have used in films with nominal thicknesses less then the grain size. To fully resolve this problem, further experimental and theoretical work is needed on the electron-phonon interaction in disordered thin films.[34] In particular, the dependence of $\tau_\phi(T)$ on morphology in ultrathin films should be further investigated.

There have been three previous studies of the MC of thin Nb films. Moehlecke and Ovadyahu[35] examined the MC of high resistance ($R_\square > 1$ kΩ) nonsuperconducting films. They were unable to fit their data to the weak localization theory, perhaps because this theory may not apply to such large $R_\square$ samples. Leeman, Wolf, Elliott, and Orbach[36] also studied the MC of nonsuperconducting Nb films. Their samples were produced by anodizing 50-Å sputtered films until $R_\square$ fell in the range 500—2000 Ω. They found $\tau_\phi^{-1} \propto T$ over the temperature range 1.4 to 4.2 K, with $\tau_\phi = 4$ psec at 4.2 K. These results are consistent with ours. A linear temperature dependence over this temperature range is expected in higher $R_\square$ films where $\tau_{e\text{-}e}^{-1}$ is more dominant. Furthermore, the magnitude of the τ_ϕ they report at 4.2 K is consistent with Eq. (4).

Gershenzon, Gubankov, and Zhuravlev[37] studied the MC of superconducting Nb films with $R_\square$ in the general range that we have investigated. They also found a negative MC at all temperatures and concluded that $\tau_\phi \gg \tau_{SO}$. For their samples, $\tau_\phi^{-1} \propto T$ at the lowest temperatures, but they found a crossover to a T^2 behavior at higher temperatures. Despite this different exponent, the magnitude of their $\tau_\phi(T)$ data is in good general agreement with ours.

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