Article

Solving mystery with the Meissner state in $La_3Ni_2O_{7-\delta}$

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Received September, 22, 2024; revised October, 13, 2024; accepted October, 16, 2024.

Recently, zero resistance state in highly compressed $La_3Ni_2O_{7-\delta}$ has been observed. However, all attempts of many research groups to detect the Meissner state in the $La_3Ni_2O_{7-\delta}$ have been failed. To explain this puzzle, an exotic superconducting state (for instance, filamentary superconductivity) in the $La_3Ni_2O_{7-\delta}$ has been supposed. Here, I extracted temperature dependent self-field critical current, $I_c(sf,T)$, dataset from current-voltage curves and performed the $I_c(sf,T)$ analysis. As a result, I found that highly compressed $La_3Ni_2O_{7-\delta}$ to exhibits *d*-wave superconductivity with the gap-to-transition temperature ratio $2\Delta(0)/(k_BT_c) = 4.0 \pm 0.3$, a very large ground state London penetration depth, $\lambda(0, P = 16.6$ GPa$) = 6.0$ μm, and a very high Ginzburg-Landau parameter $k(0, P = 16.6$ GPa$) = 1500$. This implies that the ground state lower critical field $B_{c1}(0, P = 16.6$ GPa$) = 34$ μT is of the same order as the Earth's magnetic field. Based on this, to detect the Meissner state in the $La_3Ni_2O_{7-\delta}$ becomes a very challenging task. I can hypothesize that the magnetic flux trap effect recently proposed to eliminate the diamond anvil cell (DAC) background in experiments on magnetic properties of the superconducting hydrides can also apply in studies of magnetic properties in the $La_3Ni_2O_{7-\delta}$ superconductor.

Keywords: High-pressure superconductivity; Nickelate superconductors; London penetration depth; Lower critical field.

DOI: 10.62539/2949-5644-2024-0-3-65-76

1. Introduction

High-temperature superconductivity in nickelates, predicted by Anisimov et al. [1] in 1999, has been experimentally observed in infinite layer nickelates [2] and the Ruddlesden-Popper layered nickelates [3]. Despite the $La_3Ni_2O_{7-\delta}$ and $La_4Ni_3O_{10}$ had started to be studied three decades ago [4–6], only Sun et al. [3] applied high pressure on RP nickelate and observed evidences for the $T_{c,onset} \sim 80$ K. Current status of the problem has discussed in [7–35]. For the completeness, there is a need to mention recent reports [36–38] where important results in high pressure superconductivity have been reported.

While in the first paper on the superconductivity in $La_3Ni_2O_{7-\delta}$ [3] the zero-resistance state has not been observed, recently Zhang et al. [39] reported the achieving the zero-resistance state in several $La_3Ni_2O_{7-\delta}$ single crystals subjected to the pressure within the range of $P = 13.7$–29.2 GPa. However, another unique property of all superconductors, which is the diamagnetism, has been never observed in the $La_3Ni_2O_{7-\delta}$. To explain this experimental fact, Zhou et al. [24] proposed exotic filamentary type of superconductivity in highly compressed $La_3Ni_2O_{7-\delta}$.

Here, I analysed the self-field critical current density, $J_c(sf,T)$, in the $La_3Ni_2O_{7-\delta}$ single crystals by the universal equation [40, 41]:

$$J_c(sf,T) = \frac{\phi_0}{4\pi\mu_0}\frac{ln(\kappa)+0.5}{\lambda^3(T)} \times \left(\frac{\lambda(T)}{a}\tanh\left(\frac{a}{\lambda(T)}\right) + \frac{\lambda(T)}{b}\tanh\left(\frac{b}{\lambda(T)}\right)\right), \tag{1}$$

where ϕ_0 is the superconducting magnetic flux quantum, μ_0 is the permeability of free space, $\lambda(T)$ is the London penetration depth, $\kappa = \lambda(0)/\xi(0)$ is the Ginzburg-Landau parameter, where $\xi(0)$ is the ground state coherence length, $2a$ is the sample cross-sectional width, and $2b$ is the sample thickness.

The analysis revealed that the $La_3Ni_2O_{7-\delta}$ to exhibits:

1. *d*-wave superconductivity;
2. gap-to-transition temperature ratio, $2\Delta(0)/(k_BT_c)$ (at $P = 16.6$ GPa$) = 4.0 \pm 0.3$;

3. a very large ground state London penetration depth $\lambda(0, P = 16.6$ GPa$) = 6$ μm;

4. extremely high Ginzburg-Landau parameter $\kappa(0, P = 16.6$ GPa$) = 1500$;

5. vanishingly small ground state lower critical field, B_{c1} $(0, P = 16.6$ GPa$) = 34$ μT.

These findings explain why the Meissner state has been never experimentally observed in the $La_3Ni_2O_{7-\delta}$. The reason is that any external magnetic field, similar to it amplitude to the Earth's magnetic field, destroys the Meissner state in the $La_3Ni_2O_{7-\delta}$ even at very low temperature. Even considering that the mixed state in type-II superconductors exists up to the upper critical field (this field is about $B_{c2}(0) \sim 20$ T in the $La_3Ni_2O_{7-\delta}$), to extract the diamagnetic response of this amplitude from a bulk sample subjected to high pressure is remarkably complicated experimental task.

It looks more realistic to detect and measure another fundamental magnetic response of the superconductors, which is the magnetic flux trap [42]. In these experiments, the applied magnetic field, B_{appl}, is equal to zero. The methodology for performing the flux trap experiments has been described by Minkov et al. [43] recently, for the registering the trap flux effect in highly compressed hydrides H_3S [44] and LaH_{10} [45, 46], while Bhattacharyya et al. [47] detect the effect in CeH_9 [47–50] by utilizing a different approach. Current status of the studies of in this field has been reviewed recently [51–53].

2. Self-field critical current density dataset in $La_3Ni_2O_{7-\delta}$

Voltage-current datasets, $V(I)$, measured by Zhang et al. [39] at different temperatures and pressures, were first recalculated to the $E(J)$ datasets by assuming reported [39] sample cross-sectional dimensions, $2a = 70$ μm and $2b = 5$ μm, and assumed distance of $l = 100$ μm between voltage potential tabs.

Analyzing obtained $E(J)$ datasets, we concluded that standard electric field criterion of $E_c = 1$ μV/cm [54–58] cannot be used to analyze $E(J)$ datasets of $La_3Ni_2O_{7-\delta}$, because transport current change step was relatively large of $\Delta I = 0.1$–0.2 mA, or $\Delta J = (2.85$–$5.7)\times10^5$ A/m^2.

By experimenting with different E_c criteria, I found that the electric field criterion of $E_c = 3$ mV/cm represents a suitable balance between the accuracy of experimental data and the requirement to use as low as possible E_c criterion. Despite this criterion exceeds the conventional criterion of $E_c = 1$ μV/cm by 3000 times, is by 33 times lower than the strictest criterion of $E_c = 100$ mV/cm used to define critical currents in Moiré 2D films [59].

The $E(J)$ datasets were fitted to the equation:

$$E(J) = E_0 + \frac{|J|}{J} \times E_c \times \left(\frac{J}{J_c}\right)^n, \tag{2}$$

where E_0 is the offset originating in the amplifier, J_c is the critical current density, and n is the power law exponent. Fits are shown in Fig. 1.

3. Ground state coherence length in $La_3Ni_2O_{7-\delta}$

Zhang et al. [39] reported $B_{c2}(T)$ datasets defined at $T = T_c^0$, where T_c^0 is the transition temperature defined by the strict resistance criterion:

$$\frac{R\left(T = T_c^0\right)}{R_{norm}\left(T\right)} \leq 10^{-3}, \tag{3}$$

where $R_{norm}(T)$ is the resistance at temperatures higher than the temperature at $R(T)$ starts to decrease. $B_{c2}(T)$ datasets has been by Zhang et al. [40] in their Extended Data Fig. 6 [39].

These datasets were fitted (Fig. 2) to the approximated equation of the Werthamer-Helfand-Hohenberg (WHH) [60, 61] theory, which was proposed by Baumgartner et al. [62]:

$$B_{c2}(T) = \frac{1}{0.693} \times \frac{\phi_0}{2\pi\xi^2(0)} \times \left(\left(1 - \frac{T}{T_c}\right) - 0.153 \times \left(1 - \frac{T}{T_c}\right)^2 - 0.152 \times \left(1 - \frac{T}{T_c}\right)^4 \right) \quad (4)$$

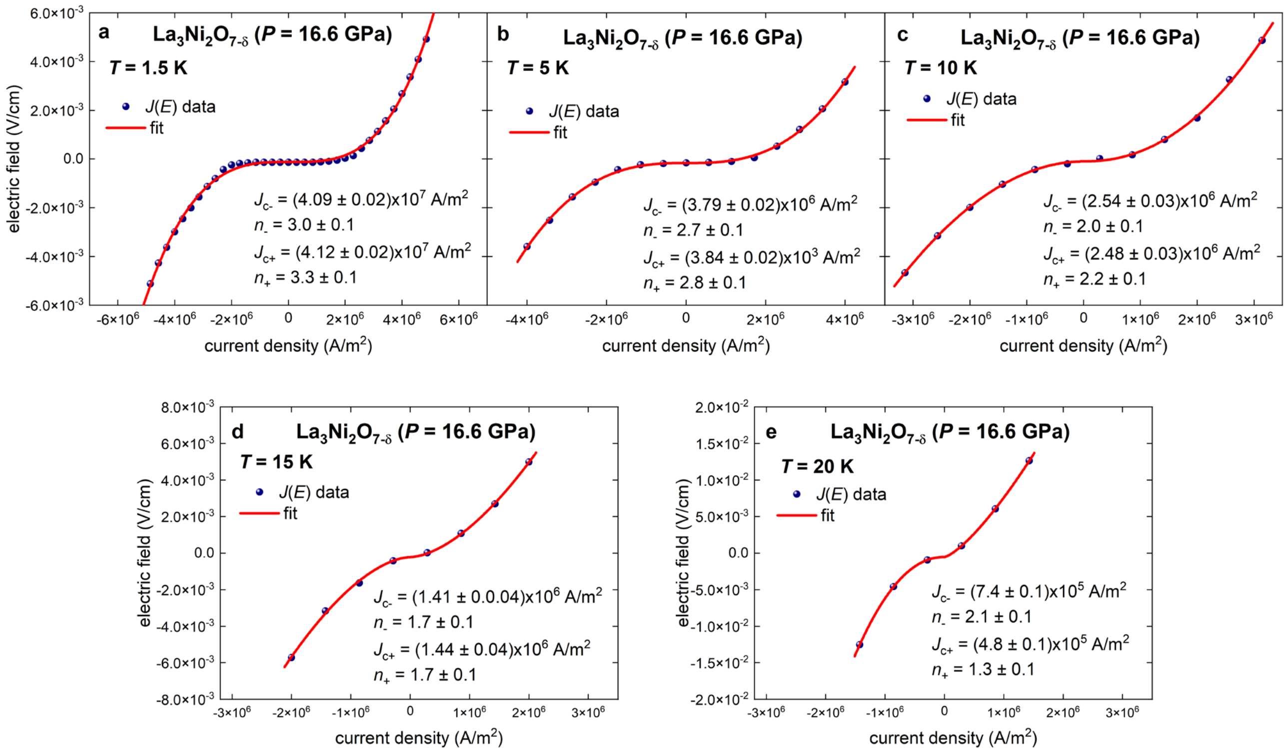

Fig. 1. $E(J)$ datasets reported by Zhang et al. [39] for highly compressed $La_3Ni_2O_{7-\delta}$ and data fits to Eq. 2. Derived self-field critical current density, $J_c(sf,T)$, and n-values are shown for each temperature.

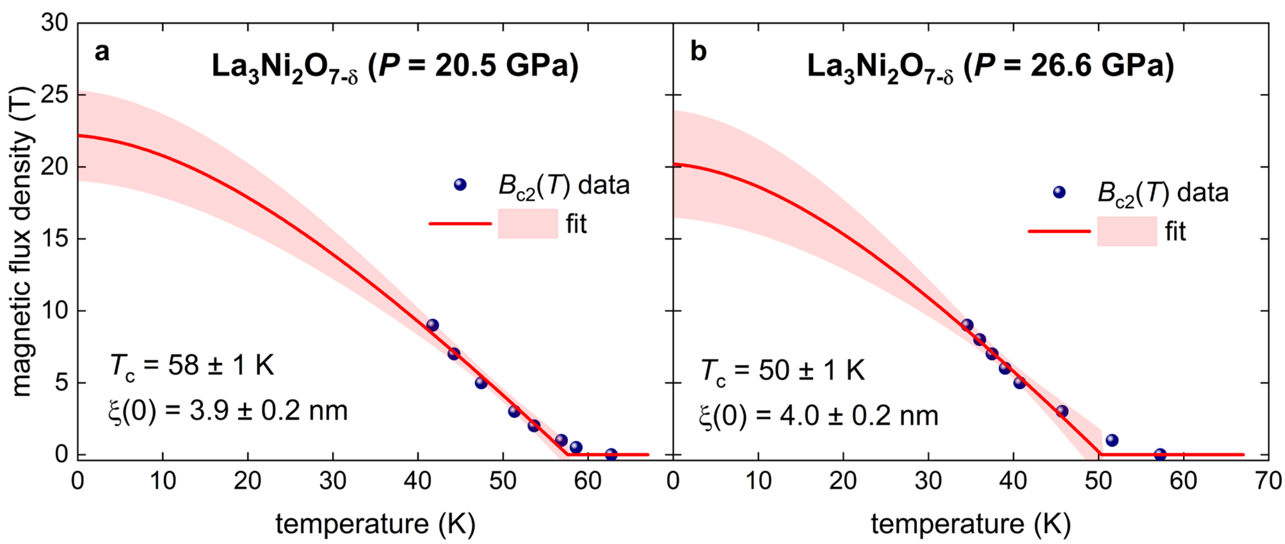

Fig. 2. $B_{c2}(T)$ datasets reported by Zhang et al. [39] for highly compressed $La_3Ni_2O_{7-\delta}$ and data fits to Eq. 4. Derived $\xi(0)$ and T_c are shown for each panel. The 95% confidence bands are shown by the pink areas.

Deduced ground state coherence length, $\xi(0)$, for $La_3Ni_2O_{7-\delta}$ compressed at $P = 20.5$ GPa and $P = 26.6$ GPa are shown in Fig. 2a and Fig. 2b respectively.

Recently, Prozorov and Kogan [63] proposed practical universal equation within the WHH formalism:

$$B_{c2}(T) = \frac{\phi_0}{2\pi\xi^2(0)} \times \left(\frac{1 - \left(\frac{T}{T_c}\right)^2}{1 + 0.42 \times \left(\frac{T}{T_c}\right)^{1.47}} \right) \tag{5}$$

$B_{c2}(T)$ data fits to Eq. 5 and deduced ground state coherence length, $\xi(0)$, for $La_3Ni_2O_{7-\delta}$ compressed at $P = 20.5$ GPa and $P = 26.6$ GPa are shown in Fig. 3a and Fig. 3b respectively.

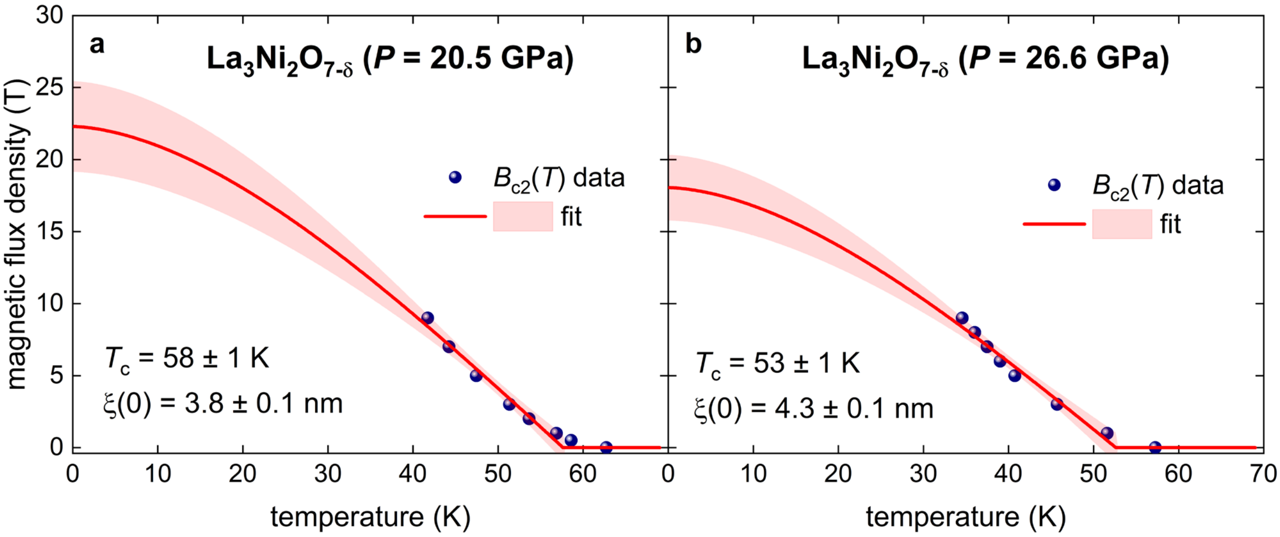

Fig. 3. $B_{c2}(T)$ datasets reported by Zhang et al. [40] for highly compressed $La_3Ni_2O_{7-\delta}$ and data fits to Eq. 5. Derived $\xi(0)$ and T_c are shown for each panel. The 95% confidence bands are shown by the pink areas.

Based on the results of performed fits showed in Figs. 2, 3, the average rounded value of $\xi(0) = 4$ nm has been assumed as the ground state coherence length in $La_3Ni_2O_{7-\delta}$. This value was substituted in Eq. 1, as a fixed parameter.

4. Fundamental superconducting parameters of the $La_3Ni_2O_{7-\delta}$

Eq. 1 was used to fit $J_c(sf,T)$ datasets measured in form of nanowires, thin films and bulk superconductors [40, 41, 48, 59, 64–86] to extract the ground state London penetration depth, $\lambda(0)$, ground state energy gap, $\Delta(0)$, and the relative jump in electronic specific heat at the transition temperature, $\Delta C_{el}/(\gamma T_c)$, where γ is the Sommerfeld constant. Despite details of the approach can be found elsewhere [40, 41] (and free-online software to extract the $\lambda(0)$, $\Delta(0)$, $\Delta C_{el}/(\gamma T_c)$, and T_c from experimental $J_c(sf,T)$ datasets is available [41]) it would be useful to make a short clarification of the method [40, 41].

In Eq. 1 temperature dependent London penetration depth for s-wave superconductors is described by the following equation:

$$\lambda(T) = \frac{\lambda(0)}{\sqrt{1 - \frac{1}{2k_BT} \cdot \int_0^\infty \frac{d\varepsilon}{\cosh^2\left(\frac{\sqrt{\varepsilon^2 + \Delta^2(T)}}{2 \cdot k_BT}\right)}}}, \tag{6}$$

where k_B is the Boltzmann constant, and temperature dependent gap amplitude is given by Gross et al. [87, 88]:

$$\Delta(T)=\Delta(0)\times\tanh\left[\frac{\pi k_B T_c}{\Delta(0)}\times\sqrt{\eta\times\frac{\Delta C_{el}}{\gamma T_c}\times\left(\frac{T_c}{T}-1\right)}\right], \quad (7)$$

where $\eta \equiv 2/3$ for s-wave superconductors [87, 88].

For d-wave superconductors, temperature dependent London penetration depth is given by [87, 88]:

$$\lambda(T)=\frac{\lambda(0)}{\sqrt{1-\frac{1}{2\pi k_B T}\cdot\int_0^{2\pi}\cos^2(\theta)\cdot\left(\int_0^{\infty}\frac{d\varepsilon}{\cosh^2\left(\frac{\sqrt{\varepsilon^2+\Delta^2(T,\theta)}}{2k_B T}\right)}\right)\cdot d\theta}}, \quad (8)$$

where the superconducting energy gap, $\Delta(T,\theta)$, is given by [87, 88]:

$$\Delta(T,\theta)=\Delta_m(T)\times\cos(2\theta), \quad (9)$$

where $\Delta_m(T)$ is the maximum amplitude of the k-dependent d-wave gap given by Eq. 7, θ is the angle around the Fermi surface subtended at (π, π) in the Brillouin zone (details can be found elsewhere [87, 88]), and $\eta \equiv 7/5$ in accordance with Refs. [87, 88].

Result of the fit of the obtained $J_c(sf,T)$ dataset measured for the $La_3Ni_2O_{7-\delta}$ sample (subjected to pressure P = 16.6 GPa) to Eq. 1 is shown in Fig. 4.

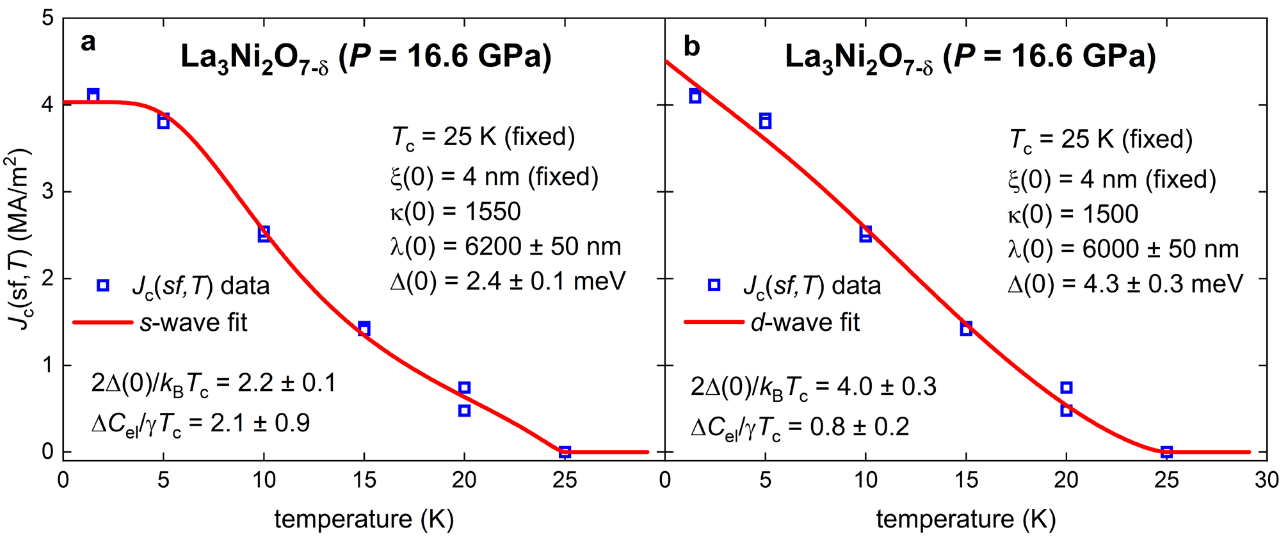

Fig. 4. $J_c(sf, T, P$ = 16.6 GPa) dataset and data fit to (a) s-wave and (b) d-wave gap symmetry. Deduced parameters are shown in each panel. Fit quality is (a) 0.9641, and (b) 0.9119.

To reduce the number of free fitting parameters, I fixed T_c = 25 K for the fit. One can see that deduced values favour the d-wave symmetry, for which the following characteristics were deduced:

1. ground state London penetration depth $\lambda(0) = 6000 \pm 50$ nm;
2. ground state energy gap $\Delta_m(0)=4.3 \pm 0.3$ meV;
3. gap-to-transition temperature ratio, $2\Delta_m(0)/(k_B T_c) = 4.0 \pm 0.3$;
4. the relative jump in electronic specific heat at the transition temperature, $\Delta C_{el}/(\gamma T_c) = 0.8 \pm 0.2$;
5. ground state Ginzburg-Landau parameter $\kappa(0) = 1500$.

Deduced parameters (Fig. 3) demonstrate that the $La_3Ni_2O_{7-\delta}$ (subjected to pressure $P = 16.5$ GPa) is an extremely high-k weak-coupled *d*-wave superconductor. In a literature [89], the designation of high-k superconductor is using for superconductors, which exhibit the Ginzburg-Landau parameter $k \geq 50$. For instance, MgB_2 (exhibited $k \approx 25$) is usually classified as type-II superconductor [90], while all HTS cuprates with $k \geq 50$ and iron-based superconductors are classified as high-k superconductors.

5. Ground state lower critical field in $La_3Ni_2O_{7-\delta}$

Deduced parameters give the answer to the main unanswered question to date: why the Meissner state is not observed in the $La_3Ni_2O_{7-\delta}$, while the zero resistance has observed in $La_3Ni_2O_{7-\delta}$. The answer is that the ground state lower critical field is remarkably low. This value is calculated by the equation:

$$B_{c1}(0, P = 16.6\,GPa) = \frac{\phi_0}{4\pi}\frac{ln(\kappa)+0.5}{\lambda^2(0)} = 34 \quad \mu T \tag{10}$$

Eq. 10 shows that even at very low temperatures, the amplitude of the lower critical field has amplitude as the Earth's magnetic field and, thus, it is remarkably difficult to detect this field in an experiment.

6. Discussion

London penetration depth, $\lambda(T)$, is one of two primary fundamental lengths of any superconductor, which is linked with the bulk density of the Cooper's pairs, $n_s(T)$, in the superconductor [42, 77, 87, 90–94]:

$$n_s(T) = \frac{m_e}{\mu_0 e^2 \lambda^2(T)}, \tag{11}$$

where m_e is the mass of the charge carrier, and e is the electron charge.

It should be mentioned that the ambient pressure organic superconductors exhibit similar $\lambda(0)$ and $B_{c1}(0)$ to the values determined for the $La_3Ni_2O_{7-\delta}$ herein. And, thus, the large London penetration depth, $\lambda(T)$, in the $La_3Ni_2O_{7-\delta}$ and in organic superconductors originates from the low bulk density of the Cooper pairs in these materials.

For instance, deuterated $(TMTSF)_2ClO_4$ exhibit macroscopically large ground state London penetration depth $\lambda_b(0) = 40$ µm [95]. In this compound $B_{c1,b}(0) = 55$ µT [95], which is not much different from derived in this study the $B_{c1,c}(0, P = 16.6\text{ GPa}) = 34$ µT in the highly compressed $La_3Ni_2O_{7-\delta}$. Another ambient pressure organic superconductor, which can be mentioned, is the $TaS_2(pyridine)_{1/2}$, which exhibits $\lambda_c(0) = 8–130$ µm [96, 97]. In this regard, well-studied the $YBa_2Cu_3O_{7-\delta}$ cuprate superconductor exhibits $\lambda_c(0, \delta = 0.3) = 4.53$ µm and $\lambda_c(0, \delta = 0.43) = 7.17$ µm [98].

To the best of my knowledge, the lowest value for the lower critical field has been reported for *b*-$(BEDT\text{-}TTF)_2I_3$. This compound exhibits [99] anisotropic fields of $B_{c1,||a}(0) = 5.2 \pm 0.5$ µT and $B_{c1,\perp a}(0) = 8.7 \pm 1.4$ µT, which all well below the derived value $B_{c1,c}(0, P = 16.6\text{ GPa}) = 34$ µT for $La_3Ni_2O_{7-\delta}$. More examples of superconductors with large London penetration depth can be found in

Refs. [91, 99, 100].

Deduced parameters for the $La_3Ni_2O_{7-\delta}$ explain why it has been extremely difficult to observe the Meissner state experimentally observed.

There is a need to discuss the influence of possible multi-band superconductivity [101, 102] in $La_3Ni_2O_{7-\delta}$ on the lower critical field, B_{c1}, and the London penetration depth, λ. As a rule, multiband superconductors are described by the so-called γ-model, where total superfluid density [103, 104], the specific heat capacity [105], critical current density [41, 65, 106], the lower critical field [107], and the upper critical field [59, 108–111] are defined as the linear sum of a contribution from each band in proportion to the partial Sommerfeld constants, γ_i. In the case of two-band superconductor, the equation for critical current density is [41, 59, 66, 106]:

$$J_c\left(sf,T\right)_{total} = J_c\left(sf,T\right)_{band1} + J_c\left(sf,T\right)_{band2} \tag{12}$$

and the equation for the lower critical field is [103, 107]:

$$B_{c1}\left(T\right)_{total} = B_{c1}\left(T\right)_{band1} + B_{c1}\left(T\right)_{band2} \tag{13}$$

It should be emphasized that experimentally measured values are the total ones, i.e. the $J_c(sf,T)_{total}$, $B_{c1}(T)_{total}$, $B_{c2}(T)_{total}$, *etc*.

Thus, $J_c(sf,\ T = 1.5\ \text{K})_{total}$ value (Fig. 4) and two other values, $B_{c1}(T = 1.5\ \text{K})_{total}$ (Eq. 10) and $\lambda(\text{T} = 1.5\ \text{K})_{total}$ (Fig. 4) (which were deduced from the $J_c(sf,\ T = 1.5\ \text{K})_{total}$) represent values which cannot be altered by the issue that the $La_3Ni_2O_{7-\delta}$ can be a multiple band superconductor.

Even if the $La_3Ni_2O_{7-\delta}$ is a multi-band superconductor, the detection of this feature would be possible in very clean crystals exhibited a very large free-mean path, l_{mfp}, of charge carriers [112] $l_{mfp} >> \xi(0) = 4$ nm. This condition is unlikely to be realized in presently available $La_3Ni_2O_{7-\delta}$ crystals, based on a fact that the $La_3Ni_2O_{7-\delta}$ exhibits a very large crystalline strain [113], $\varepsilon > 0.01$, which originates from external pressure and defects.

It should be noted, that recently Seshita et al. [114] reported $\lambda(0) = 3.2$ μm in high-entropy-type (HE-type) superconductor $AgInSnPbBiTe_5$. Thus, large $\lambda(0)$ values (similar to the derived $\lambda(0) = 6$ μm in $La_3Ni_2O_{7-\delta}$ herein) are not unusual, and these values originate from low Copper pair density in these superconductors.

7. Conclusions

In this paper, I determined that presently available single crystals of the $La_3Ni_2O_{7-\delta}$ exhibit very large London penetration depth, $\lambda(0) = 6$ μm and, as a direct consequence of this, small ground state lower critical field $B_{c1}(0) = 34$ μT.

The best option for observing of the unique magnetic response of the $La_3Ni_2O_{7-\delta}$ superconductor is to perform the flux trap effect [42] measurements. This technique was recently successfully implemented by Minkov et al. [43] in studies of highly compressed hydrides.

Acknowledgements

The author thanks for the financial support provided by the Ministry of Science and Higher Education of Russia (theme “Pressure” No. 122021000032-5).

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УДК 538.945

Разгадка тайны мейсснеровского состояния в $La_3Ni_2O_{7-\delta}$

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Недавно было обнаружено состояние нулевого сопротивления в сильно сжатом $La_3Ni_2O_{7-\delta}$. Однако все попытки многих исследовательских групп обнаружить состояние Мейсснера в $La_3Ni_2O_{7-\delta}$ потерпели неудачу. Чтобы объяснить экспериментальные факты, несколько экзотических видов сверхпроводимости было предложено для $La_3Ni_2O_{7-\delta}$, и в частности состояние, названное нитевидной сверхпроводимостью. В данной работе я обработал вольт-амперные кривые и определил температурную зависимость критического тока в собственном поле, $I_c(sf,T)$. Анализ полученной экспериментальной зависимости $I_c(sf,T)$ показал, что сильно сжатый $La_3Ni_2O_{7-\delta}$ имеет сверхпроводимость с симметрией энергетической щели по типу *d*-wave, с отношением щели к температуре перехода $2\Delta(0)/(k_BT_c) = 4.0 \pm 0.3$, очень большой глубиной проникновения Лондона в основном состоянии, $\lambda(0, P = 16.6$ ГПа$) = 6.0$ мкм, а также очень высоким параметром Гинзбурга-Ландау $k(0, P = 16.6$ ГПа$) = 1500$. Эти величины показывают, что нижнее критическое поле основного состояния $B_{c1}(0, P = 16.6$ ГПа$) = 34$ мкТл, и практически означает, что $B_{c1}(0)$ имеет тот же порядок, что и магнитное поле Земли. Исходя из этого, обнаружение состояния Мейсснера в $La_3Ni_2O_{7-\delta}$ представляет собой очень сложную задачу. Я могу выдвинуть гипотезу, что эффект захвата магнитного потока, недавно использованный для устранения фонового сигнала ячейки с алмазными наковальнями (DAC) в экспериментах по магнитным свойствам сверхпроводящих гидридов, может быть также применен в исследованиях сверхпроводника $La_3Ni_2O_{7-\delta}$.

Ключевые слова: Сверхпроводимость в материалах при экстермальных условиях; сверхпроводимость в оксидах никеля; лондоновская глубина проникновения; нижнее критическое поле.

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