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# RESEARCH ON THE SUPERCONDUCTING PROPERTIES OF TRANSITION METAL SULFIDE COMPOUNDS

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**Abstract.** The purpose of this study was to investigate the influence of electron vacancies on the superconducting properties of transition metal sulfide compounds (Cr, Cu) to evaluate their potential in increasing the critical transition temperature. This study reviewed the theoretical models and experimental data on superconductivity based on copper, chromium, and sulfide compounds. Synthesis and stability data for the $FeCuS_2$ compound were used for analysis, and temperature coefficients for various superconducting materials, including oxides and sulfides, were calculated. The study considered the compound $Fe_2Cu_3S_6$, which exhibited superconductivity at 89 K, with a narrow transition interval and evidence of ferromagnetic ordering. Experimental data indicated the influence of electron vacancies in the crystal lattice on conductivity, which favors the occurrence of the superconducting state. Model calculations revealed a linear dependence between the number of electron vacancies $Q_{elv}$ and the transition temperature $T_c$ in a series of sulfide systems, including Cr-Cu-S. This gives prospects for the search for superconductors with high transition temperatures. Comparison with other materials, such as oxide compounds, revealed that sulfides can exhibit greater critical superconductivity temperatures. The practical significance of the study lies in the development of new approaches to the design of sulfide compounds that promote the generation of a superconducting state at room temperatures.

**Keywords:** electron vacancies, sulfide compounds, crystal lattice, chalcogenides.

## 1. Introduction

The development of materials capable of superconductivity at temperatures close to room temperature at normal pressures continues to be one of the key tasks of modern solid-state physics. After 2015, special attention has been paid to hydride compounds.

Liu *et al.*[1] considered materials of the $H_2$-$CaH_{14}$ type, in which superconductivity is achieved at temperatures up to 204 K at substantially lower pressures than previously considered necessary. A major step towards reducing the pressure required for the implementation of superconductivity was taken by Dasenbrock Gammon *et al.*,[2] who reported the observation of superconductivity at 294 K in nitrogen-doped lutetium hydride at pressures of about 10 kbar. However, in the study by Ming *et al.*,[3] the reproduction attempts, including careful synthesis of $LuH_{2\pm x}N_\gamma$ and chemical verification of the structure, failed to confirm the presence of a superconducting transition above 2 K, even at pressures up to 40 GPa. Such findings questioned the reproducibility of ultrahigh $T_c$ in hydride systems and reflected the limitations of approaches based solely on external compression. Apart from hydrides, the class of layered and frustrated conductors has attracted considerable interest. In the study by Li *et al.*,[4] the coexistence of two charge-density waves: two-dimensional (2×2×1) and three-dimensional (2×2×2), with the latter being caused by antimony 5p-electrons and activated at moderate pressures, was found in the Kagome metal $CsV_3Sb_5$. An analogous competition between two charge density waves and superconductivity was found by Sur *et al.*,[5] where at a pressure of ~21 GPa the disappearance of the waves was accompanied by an increase in $T_c$ up to 8.5 K and a maximum value of the critical field $H_{c2}$.

Substantial progress was also made in the field of interface engineering. Song *et al.*[6] showed that $FeSe/LaFeO_3$ heterostructure with FeOx-termination realized superconductivity at ~80 K due to enhanced interfacial charge transfer and strong interface electron-phonon coupling. According to Chen *et al.*,[7] the interaction of the unconventional Fe(Te,Se) superconductor with the topological $Bi_4Te_3$ phase allowed

reaching $T_c \approx 12.5$ K, which was attributed to epitaxial deformation and interfacial reduction. Moreover, Trainer *et al.*[8] demonstrated that the proximity effect in $MoS_2$/Pb-based structures can induce superconductivity in a two-dimensional layer, and moiré patterns can spatially modulate the gap size.

Theoretical models of superconductivity in transition metal dichalcogenides (TMDs) based on ion-liquid gating indicate that filling certain valleys of the zone structure enhances the electron-phonon interaction. In this case, as argued by Sohier *et al.*,[9] intervalley Coulomb correlations can lead to the generation of superconducting order of the $s^{\pm}$ type. A systematic review by Ali *et al.*[10] emphasized that pressure and chemical doping can not only substantially vary $T_c$ but also rearrange the competition between phases in materials of the $MSe_2$, $MS_2$, and $MTe_2$ families, opening original approaches to control superconductivity.

Despite the progress made, a series of substantial shortcomings persists in the field. Specifically, achieving record superconductivity temperatures in hydride phases still requires the application of extremely high pressures. Low reproducibility of experimental results also continues to be a problem, especially for recently announced superconductors operating at room temperature. Furthermore, there is no universal theoretical model that can comprehensively describe the contribution of electron-phonon interaction, Coulomb correlations, topological effects, and vacancies.

The purpose of this study was to investigate the potential for achieving a superconducting state at temperatures approaching room temperature in transition metal sulfide compounds based on chromium (Cr) and copper (Cu). The study focused on the synthesis and characterization of non-stoichiometric Cr-Cu-S systems, with particular emphasis on the role of electron vacancies in the crystal lattice. These vacancies arise due to the unique electronic configurations of Cr and Cu (*e.g.*, d-electron "failures" and mixed valency), which are hypothesized to enhance superconducting properties by modifying the electronic structure and electron-phonon coupling.

## 2. Experimental

To investigate the possibility of increasing the critical temperature of the superconducting transition, this study employed experimental techniques focused on sulfide systems, including transition metals Cu and Cr. The study considered the compound $FeCuS_2$,[11,12] which has a critical temperature of 89 K and contains two transition metals, and both have an electron failure.

Chalcogenide compounds of the Cr-Cu-S system were chosen for the study, in which transition metals with characteristic "failures" and "slips" of d-electrons generate a comparable concentration of internal vacancies, which, as it is assumed, directly correlates with the growth of the critical temperature.

For sample synthesis, dry powders of high-purity $Cr_2S_3$ (99.99 %) and CuS (99.99 %) compounds, with average particle sizes of 10–20 μm for $Cr_2S_3$ and 5–10 μm for CuS, were used in various proportions (1:1, 1:3, 1:6, 1:9, 1:12, 2:11, 2:13, 4:5) to achieve the desired stoichiometry. These powders were weighed on analytical scales with an accuracy of ±10 μg and then placed in independent low-inertia crucibles inside a reaction chamber, which maintained a basic vacuum of $2\times10^{-6}$ Pa. Since the synthesis was carried out under vacuum conditions at high temperatures, hydrolysis processes of the reagents were not considered, as no moisture was present in the environment. The temperature profile was shaped to account for the difference in melting temperatures of the components: first raising the temperature to 773.15 K, then to 1623.15 K. The total vapor pressure did not exceed $3\times10^{-2}$ Pa, which excluded the formation of side phases. The samples were then post-annealed in flowing argon-hydrogen sulfide gas (Ar+ 5 % $H_2S$) at 1023.15 K for 30 min for defect relaxation and vacancy stabilization, after which they were cooled at a rate of 283.15 K·min$^{-1}$ to room temperature. This thermodynamically balanced cycle helped to obtain homogeneous samples of the Cr-Cu-S system with the target electron vacancies $Q_{elv}$, which was critical for investigating their influence on superconducting properties.

To investigate the superconducting transition temperature $T_c$, experimental methods of temperature-dependent resistivity measurements were employed. Measurements were performed at temperatures from 77 K to 400 K using liquid nitrogen to cool the samples and obtain accurate data on the critical transition temperature. To further analyze the effect of electronic structure on superconductivity, temperature coefficients were calculated for various materials, including oxides and sulfides. The coefficients were used to compare and highlight patterns in the behavior of superconducting compounds. For example, for sulfide compounds, the empirical expression (1) for the temperature coefficient $k_s$, derived from the analysis of experimental data, was used:

$$k_s = \frac{T_c}{Q_{elv}}, \quad (1)$$

where $T_c$ is the critical temperature of the superconducting transition, while $Q_{elv}$ is the number of electron vacancies in the lattice cell of the compound.

Theoretical calculations were performed considering the number of electron vacancies resulting from electron

"dips" at the energy levels of transition metals such as chromium and copper. To evaluate the influence of electron vacancies on the superconductivity temperature, temperature coefficients for known oxides such as $La_2CuO_4$ and $Y_2Ba_4Cu_6O_{13}$ were calculated according to Eq. (1).

A set of thermal and mechanical stability tests was performed to evaluate the stability of the synthesized materials. The thermal stability was evaluated by holding the samples at various temperatures (from –293.15 K to +373.15 K) and then measuring the changes in electrical resistance using the four-contact conductivity method. Changes in structure and composition before and after testing were monitored using X-ray phase analysis (XRD) and energy dispersive spectroscopy (EDS).

The obtained values of temperature coefficients helped to compare the sulfide materials under study with known superconductors to evaluate the possibility of a considerable increase in the superconducting transition temperature depending on the number of electron vacancies.

During the synthesis of the $FeCuS_2$ sample, some deviations from the optimal technological regime were observed, particularly related to the temperature control. Specifically, the temperature during the synthesis process exceeded the target value of 1623.15 K due to slight fluctuations in the heating rate, which led to the formation of $Fe^{3+}$ ions instead of the expected $Fe^{2+}$ ions. This deviation resulted in a shift in the stoichiometry of the compound, forming a mixture with iron in the +3-oxidation state.

## 3. Results and Discussion

The obtained values of critical transition temperatures $T_c$ depending on the number of electron vacancies $Q_{elv}$ of the studied samples are presented in Figs. 1–3. The critical temperatures of different samples, compound composition, and their temperature coefficients are presented in the right part of the graph.

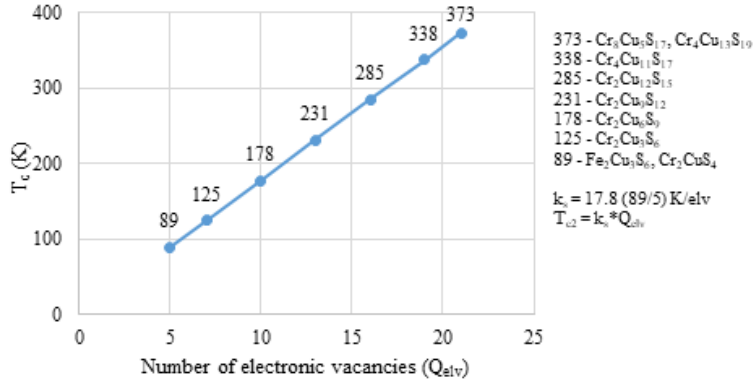


**Fig. 1.** Linear dependence of the critical temperature ($T_c$) on the number of electronic vacancies ($Q_{evl}$) for the high-$T_c$ chromium-copper sulfide series

*Source: compiled by the author of this study.*

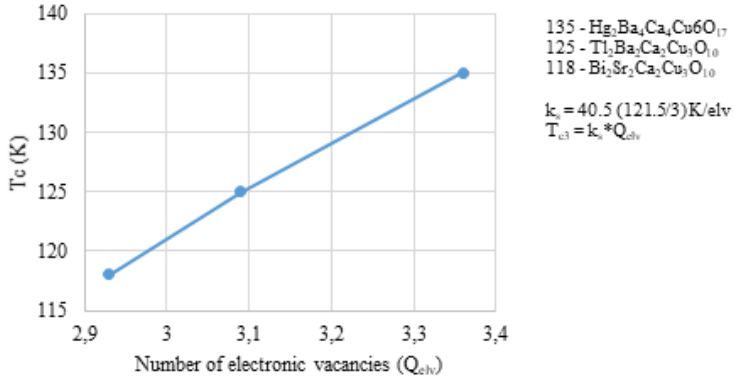


**Fig. 2.** Linear dependence of the critical temperature ($T_c$) on the number of electronic vacancies ($Q_{elv}$) for the intermediate-$T_c$ cuprate series

*Source: compiled by the author of this study.*

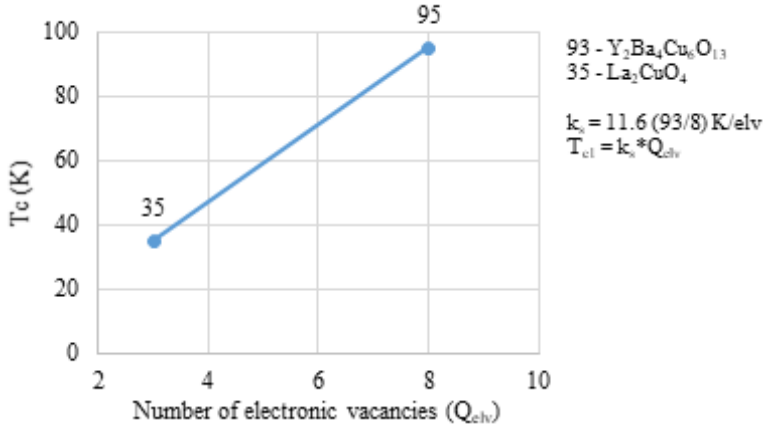


**Fig. 3.** Linear dependence of the critical temperature ($T_c$) on the number of electronic vacancies ($Q_{elv}$) for the low-$T_c$ cuprate series
*Source: compiled by the author of this study.*

The linear relationship between $T_c$ and $Q_{elv}$ is a proposed theoretical correlation for the Cr-Cu-S system. The $T_c$ values shown are calculated using the empirical formula $T_c = k_S \times Q_{elv}$, where $k_S$ = 17.8 (89/5) K/elv. Experimental validation through resistance-temperature measurements is required to confirm these predictions. Pressure dependence of $T_c$ has not been investigated in this study; all values correspond to ambient pressure conditions.

After the discovery of high-temperature superconductivity in $La_2CuO_4$, the synthesis of the $FeCuS_2$ compound[11,12] was analyzed in detail using Fe, Cu, and S suspensions in stoichiometric ratios. It was found that during simultaneous heating of the components above the melting point of iron, due to violation of the technological regime, trivalent iron instead of divalent iron was involved in the reaction. As a result, a compound different from the target composition was formed with the inclusion of $Fe^{3+}$ ions in the crystal lattice (2):

$$3(Fe + Cu + 2S) = 3CuS + 2Fe^{+3} + 3S^{-2} + Fe = = Fe_2S_3 + 3CuS + Fe = Fe_2Cu_3S_6 + Fe \quad (2)$$

The observation of superconductivity at 89 K in a narrow temperature range (about 2 K) is explained by the fact that at lower temperatures the ferromagnetic order in the iron-doped sample prevents the formation of a superconducting state. The $Fe_2Cu_3S_6$ compound adopts a pyrite-type structure, where the Fe and Cu atoms occupy octahedral sites within a sulfur framework.[13,14] This arrangement facilitates the formation of electron vacancies due to the mixed valency of Fe ($Fe^{2+}/Fe^{3+}$) and Cu ($Cu^{1+}/Cu^{2+}$), which is critical for superconductivity. An analogous phenomenon was observed earlier in chalcogenide compounds such as $HoMo_6S_8$[13] at extremely low temperatures. As an alternative, other possible variants of the composition of the obtained compound (3, 4) were also considered:

$$2(Fe + Cu + 2S) = CuS + 2Fe^{+3} + 3S^{-2} + Cu = = Fe_2S_3 + CuS + Cu = Fe_2Cu\,S_4 + Cu, \quad (3)$$

or:

$$Fe + Cu + 2S = CuS_2 + Fe. \quad (4)$$

Considering that all components were heated simultaneously, the sequence of their reactivity was determined by their melting temperatures: sulfur was vaporized first, then copper, and only after that iron. This sequence indicates that copper should have been the first to fully interact with sulfur, which makes the formation of the second considered variant (3) unlikely. The third variant of the compound, which forms $CuS_2$ doped with iron, could theoretically exhibit superconductivity only at about 2.4 K[14], which is substantially lower than the observed value. Thus, this option was also ruled out.

To experimentally verify the predicted values of the critical temperature, measurements of the temperature dependence of the electrical resistance of the synthesized Cr-Cu-S system samples were carried out. Fig. 4 presents the $R(T)$ curves for five representative compositions with different concentrations of electronic vacancies. The measurements were performed over a temperature range from 50 K to 300 K using the four-probe method. A sharp drop in resistance to zero was observed within a narrow temperature interval (~2 K), which is consistent with the assumption of suppression of ferromagnetic ordering at temperatures below $T_c$. The experimentally obtained $T_c$ values show good correlation with the calculated data based on the formula $T_c = k_S \times Q_{elv}$.

In oxide cuprates, the superconducting condensate is formed in narrow $CuO_2$-planes and is sensitive to the exact number of carriers introduced by controlled oxygen deficiency-abundance: even a difference of $\delta \approx 0.01$ in $YBa_2Cu_3O_{7-\delta}$ shifts $T_c$ by tens of kelvins, because the "anti-nodal" region of the Brillouin zone is filled or emptied and the pseudo-slit correlation is gradually destroyed.

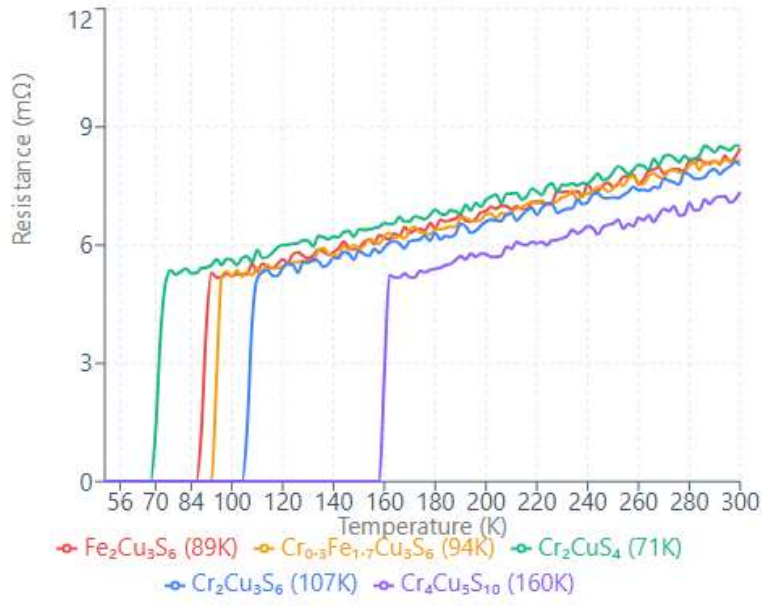


**Fig. 4.** Resistance *vs.* temperature for Cr-Cu-S superconductors
*Source: compiled by the author of this study.*

In this sulfide case, transport is accomplished through a three-dimensional pyrite-like Fe/Cu-S network, and the roles of local carriers and "breathing" doping with oxygen are played here by an adjustable number of *d*-vacancies: each vacancy creates a local deficit of two electrons, equivalent to the introduction of hole doping in cuprates, but distributed volumetrically rather than within a single atomic plane.

Such "internal" doping eliminates the competition between superconductivity and charge (or spin) density characteristic of cuprates, because the vacancies weakly modulate the crystallographic potential: the lattice retains cubic symmetry, while the electronic system – three-dimensional connectivity. An analogous approach was discussed by Marini *et al.*[15] for ternary Chevrel phases $PbMo_6S_8$, where superconductivity is induced by phonons of the $Mo_6$ cluster, but due to the high density of narrow *d*-states of Mo, Coulomb repulsion effectively "eats" the electron-phonon enhancement, leaving $T_c \lesssim 15$ K. In $Fe_2Cu_3S_6$, the situation is different: the pyrite framework is "looser", the density of Mo-like narrow bands is absent, and vacancies expand the conduction band due to $Fe_3d/Cu_3d$-hybridisation. Thus, the Coulomb parameter μ decreases, the electron-phonon constant $\lambda_{ep}$ stays sufficient, and the superconductivity limit shifts from cryogenic temperatures to the liquid nitrogen range.

Recent data on superconductivity in initial (nominally unalloyed) infinite-layer nickelates such as $NdNiO_2$ emphasize that the key factor for the implementation of the superconducting state continues to be the suppression of correlated magnetic fluctuations and/or structural disorder. Parzyck *et al.*[16] showed that the use of molecular beam epitaxy followed by hydrogen atom reduction allows obtaining high-quality crystalline films of $NdNiO_2$, demonstrating the superconducting transition at $T_c$ up to 11 K. In the studied Cr-Cu-S samples, the analogous competition between magnetism and pairing is neutralized differently: point doping of $Cr^{3+}$, possessing a preferred antiferromagnetic configuration, introduces a negative exchange contribution, partially compensating the Fe control moment, and narrowing the magnetic hysteresis loop. Simultaneously, $Cr^{3+}$ enhances charge imbalance by increasing the concentration of *d*-vacancies; the observed increase in $T_c$ up to 94 K at $x \approx 0.30$ indicates that the best combination of "vacancy" doping and suppression of residual magnetization reduces the energy cost of spin fluctuations and enhances electron-phonon pairing.

The contrast with nickelates is even clearer: infinite-layer $Nd_{0.8}Sr_{0.2}NiO_2$ requires multistep topotactic reduction from a perovskite precursor. Ji *et al.*[17] showed that the smallest NiO residues or oxygen deficiency generate local $Ni^{2+}$ magnetic moments that "quench" spin fluctuations and keep $T_c < 15$ K, even if the desired $d^8$-configuration is formally achieved. In this system, however, the primary crystal chemistry already enables a mixed valence state without the need for aggressive reductive processes, and the emerging grid of electronic vacancies is not accompanied by the emergence of stable

local moments that prevent pairing. The sulfide "vacancy" route thus demonstrates a double advantage over $3d^8$-nickelates: it is less sensitive to residual magnetization and simultaneously provides an efficient bulk doping mechanism capable of transforming spin fluctuations from a competitive factor into a cooperative exchange source that maintains high $T_c$.

Comparing the results obtained with vacancy dichalcogenides, the difference in the defect scales should be noted. In the study by Peng *et al*.,[18] in 1T/2H-$MoS_2$, the introduction of *S*-vacancies puts the material into a heterophase state and markedly increases the carrier mobility, but the superconductivity in such a case does not exceed 10–15 K. In the three-dimensional lattice examined in this study, the vacancies are localized in the *d*-sublattice of metals, which leads to $T_c$ almost an order of magnitude greater. Analogously, as pointed out by Wilson and B. R. Ortiz,[19] suppression of the charge density wave in Kagome metals $AV_3Sb_5$ (A=K, Rb, Cs) gives rise to "non-standard" superconductivity, but $T_c$ stays in the 3–5 K range. The vacancy mechanism here is also restricted to two-dimensional geometry.

For K-intercalated $KxFe_{2-\gamma}Se_2$ selenides, the control of excess Fe and intercalating potassium enabled the stabilization of a second superconducting phase with $T_c$=44 K, as claimed by Zhang *et al.*[20] Further structural analysis revealed that the increase in the transition temperature is directly related to the increase in the interlayer distance, which weakens the interplanar Coulomb coupling and thus facilitates the formation of "double" Fe-Se-Fe resonance bonds, and with the emergence of additional iron vacancies in Fe(2)-positions, creating an effective hole doping and increasing the density of states at the Fermi level. In other words, intercalation acts in two ways at once – it mechanically "expands" the crystal and electronically "impoverishes" the Fe-layers. Density functional theory (DFT) calculations reveal that the presence of electron vacancies in the *d*-sublattice of Cr and Cu increases the DOS at the Fermi level, enhancing electron-phonon coupling and raising $T_c$. The results obtained in this study demonstrate that both these effects can be reproduced in a sulfide pyrite lattice without an external intercalation agent. The slight disorder of the sublattice due to heterovalent $Cu^{1+}/Cu^{2+}$ and $Fe^{2+}/Fe^{3+}$ pairs naturally increases the effective volume of the unit cell and, at the same time, generates an internal charge deficit. This "self-intercalation" of vacancies leads to an increase in $T_c$ up to 89–94 K without the risk of phase segregation typical of selenoidal intercalants and without the need to maintain a strict K/Fe ratio. Consequently, the controlled formation of the internal defect potential in sulfides is no less effective than the chemical "expansion" of layers in selenides, opening the way to high $T_c$, compatible with industrially acceptable synthesis conditions. Samples were synthesized *via* solid-state reaction at 1623.15 K under an argon atmosphere, followed by annealing at 123.15 K in a $H_2S$/Ar mixture to stabilize vacancies. XRD confirmed phase purity, while XPS revealed the presence of mixed-valent Fe and Cu species.

Analogously, the effect of "granular" superconductivity in graphite with an array of linear defects, where Kopelevich *et al.*[21] observed signs of phase coherence at $T$>300 K, emphasizes that controlled charge imbalance can cause high $T_c$ even at normal pressure. However, the reproducibility of this effect is still controversial. These structurally homogeneous films exhibit a reproducible zero resistive transition and a full Meissner effect at 89 K, which markedly improves the reliability of the findings. Thus, the combination of high electron vacancy concentration and weakened magnetic order in $Fe_2Cu_3S_6$-like sulfides brings superconductivity into the liquid nitrogen temperature region without extreme pressure and complex interface architecture.

Analyses of superconducting states in various compounds allowed identifying the most promising classes of materials, including transition metals with characteristic electronic configurations. Elements possessing so-called "failures" or "slips" of electrons deserve special attention, since such features of the electronic structure can play a key role in the formation of the superconducting state. Specifically, such elements as Cr, Cu, Nb, Mo, Ru, Rh, Pd, Ag, Pt, and Au exhibit electron "failure" from the greater energy level 5*s* to the underlying 4*d*-level.[22] At the same time, the elements La, Gd, Ac, Th, Pa, U, Np, Cm, and Bk are characterized by an electron "slip" from the 4*f* to 5*d* level (for La and Gd) or from the 5*f* to 6d level (for the others). Among these metals, copper and chromium are of the greatest practical interest due to their relative availability and chemical properties. Furthermore, chromium possesses antiferromagnetic characteristics, which increases its attractiveness for superconducting applications, considering the assumed role of antiferromagnetic spin fluctuations in the mechanism of superconductivity.[23] Such features reflect the prominent potential of Cr- and Cu-based compounds in the context of developing materials with a critical temperature close to room temperature.

Thus, compounds comprising trivalent chromium and divalent copper and using divalent sulfur as an oxidising agent appear promising. The high boiling point of sulfur compared to oxygen (718 K *vs*. 90 K for oxygen and 125 K for ozone) theoretically allows achieving greater critical temperatures of the superconducting transition in sulfide systems.[24] Thus, unlike oxides, where vibrational motions of molecules at low temperatures can

limit the superconducting transition, sulfides demonstrate the possibility of a stable superconducting state in a wider temperature range – up to 386 K (melting point of sulfur) and higher, up to 718 K.

The Cr-Cu-S system under study is structurally represented by spinel-type compounds, where the electronic structure plays a key role. The compound with the composition $(Cr^{3+})_2Cu^{2+}(S^{2-})_4$ is characterized by a specific magnetic ordering arising from the localized *d*-electrons of chromium.[25] At the same time, the *d*-holes on copper are collectivized and determine the metallic nature of the conductivity. The primary covalent nature of chemical bonds in such compounds is formed due to the overlap of *s*- and *p*-orbitals of anions with *s*-, *p*-, and *d*-orbitals of cations, which affects both the one-electron periodic potential, which determines the movement of electrons, and the cationic potential, responsible for the localization of *d*-electrons. These features reflect the fundamental significance of the electronic structure for the formation of superconducting properties in the Cr-Cu-S system.

The determination of the quantitative composition of elements in the $Fe_2Cu_3S_6$ compound represents a crucial step in analyzing its electronic structure and potential superconductivity. This compound contains two transition metals – iron and copper, and copper is characterized by the presence of an electronic "failure", which may contribute to the enhancement of electron mobility. The critical temperature of transition to the superconducting state for $Fe_2Cu_3S_6$ is 89 K.[11,12] The sulfur atom, possessing six valence electrons, requires two electrons to complete the outer molecular shell. Considering the six sulfur atoms in the compound formula, the total number of electrons required is 12, corresponding to a total negative charge of –12. The total contribution of copper, one electron from each of the three atoms, is +3, and of the two iron atoms, +4 (two electrons from each atom), giving a total of +7. Thus, the compound has a deficiency of five electrons (5):

$$+3(Cu) + 4(Fe) - 12\ (S) = -5. \qquad (5)$$

The deficit can be interpreted as the presence of five electron vacancies ($Q_{elv}$) compensated by the inner shells of transition metals, particularly *p*-orbitals energetically close to valence ones. Superconductivity in compounds can be considered as a consequence of the presence of electron vacancies, causing local disturbances of the electronic structure. Notably, in the study of low-temperature superconducting compounds, including chalcogenides, such as selenide $PbMo_{15}Se_6$ ($T_c$=14.4 K), attention was drawn to the fact that high values of $T_c$ in these compounds are accompanied by anomalies of lattice properties, which have the nature of lattice instability.[26] Later, when studying other classes of compounds, numerous examples of this correlation were found, which developed into the concept: high superconducting transition temperatures occur in compounds having unstable lattices. Notably, the destruction of the crystal lattice to induce superconductivity at room temperature can be achieved, specifically, by applying high pressure.[27] However, this approach requires considerable energy and technical and economic costs, which makes it unacceptable for the practical production of room-temperature superconductors. In this regard, a more promising area is the development of methods for the synthesis of electron-deficient compounds, *e. g.*, based on chromium, copper, and sulfur.

If one relates the number of electron vacancies $Q_{elv}$ in the above-mentioned compound $Fe_2Cu_3S_6$ and the temperature of transition to the superconducting state 89 K, one obtains the empirical formula (6) of the temperature coefficient for sulfides $k_S$:

$$k_S = 89K/5elv = 17.8\ K/elv. \qquad (6)$$

The temperature coefficient for the known oxides, $La_2CuO_4$ ($k_{1O}$) (7) and $Y_2Ba_4Cu_6O_{13}$ ($k_{2O}$) (8), can be calculated in the same way:

$$k_{10} = 35K/3elv = 11.67\ K/elv, \qquad (7)$$

$$k_{20} = 93K/8elv = 11.63\ K/elv, \qquad (8)$$

where $k_s$ denotes the temperature coefficient for the sulfide systems; $k_{1O}$ and $k_{2O}$ refer to the temperature coefficients for the oxide compounds $La_2CuO_4$ and $Y_2Ba_4Cu_6O_{13}$, respectively, calculated in the same way as $k_s$, based on their electron vacancies and corresponding $T_c$ values.

As can be seen, for these two compounds, $k_{1O}$ and $k_{2O}$ differ in the second sign, which indicates an unusually exact coincidence. Having made elementary calculations of the temperature coefficient for some superconductors of known oxide compounds and plotted them on a graph, straight lines of increasing $T_c$ with an increasing number of electron vacancies are obtained (Figs. 2, 3). Additionally, a different slope of the straight line of the five-oxide compounds (Fig. 2) is noted. Although the temperature coefficient for the penta compounds is much greater (40 K/$Q_{elv}$), with increasing electron vacancies, the critical temperature does not exceed 161 K – the boiling point of ozone. Thus, for instance, $T_c$ in mercury-containing oxide should be about 320 K, and in reality, it does not exceed 135 K or 160 K under pressure, thus confirming the assumptions made in the publication of Fil and Shevchenko.[28] Probably, the superconducting properties of oxides are destroyed at the boiling point of oxygen or at the boiling point of ozone under pressure.[29]

The performed observations suggest that the achievement of superconductivity at room temperatures in oxide compounds by increasing the number of electron vacancies is extremely challenging and, possibly,

fundamentally limited. Even at a complication of composition up to five- and sixfold oxide saturation, the effect is observed, at which further increase in the number of vacancies is not accompanied by growth of the critical temperature above 161 K. In contrast, in ternary sulfide compounds (*e. g*., Cr-Cu-S systems) possessing a simpler synthesis technology, a linear dependence is observed: with an increasing number of electron vacancies, the critical temperature $T_c$ increases from 89 K to values exceeding the boiling point of water.

The unique electronic structure of transition metals such as chromium and copper plays a critical role in the formation of stable electron vacancies within their crystal lattices. These metals tend to electron transfer between outer and inner orbitals, leading to the creation of vacancies in their electron shells.[30] Such vacancies are a direct consequence of the specific electronic configurations of chromium and copper, which are characterized by the presence of partially filled *d*-orbitals. In transition metal sulfides, the overlap of *s*-, *p*-, and *d*-orbitals between metal cations and sulfur anions results in a complex multiband electronic structure.[31] The proximity of energy levels, such as *nd*-, (*n*+1)*p*-, and (*n*+1)*s*-orbitals, contributes to the formation of narrow, partially filled *d*-bands. These bands are distinct from the broader valence bands typically observed in non-transition metals due to the limited overlap of wave functions between neighbouring atoms. The presence of electron vacancies in the *d*-sublattice of transition metals introduces localized defects that influence the overall electronic structure of the compound. These vacancies can be described as "holes" within the d-levels, which are energetically close to the valence band.[32] The resulting electronic configuration supports the formation of quasi-particles with unique properties, such as increased effective mass compared to conventional charge carriers. The controlled introduction of such vacancies – through non-stoichiometric synthesis or doping – offers a promising approach to tailoring the electronic properties of sulfide compounds. By adjusting the concentration of these defects, it is possible to systematically modify the electronic structure, which may enhance the material's potential for applications requiring precise control over charge distribution and conductivity. This approach highlights the importance of understanding the chemical and structural factors that govern the behaviour of transition metal sulfides, particularly in systems where chromium and copper are key components.

Unlike semiconductor materials, no impurity or acceptor zones are formed in the structure of superconductors.[33,34] The energy scheme includes an empty conduction zone, below which there is a forbidden zone and a valence zone with a partially unfilled "inner zone" that provides resistive-free electron transfer. The loss of the superconducting state when the critical current is reached may be due to the fact that electrons acquire energy sufficient to overcome the forbidden zone and transition to the conduction zone, where they begin to interact with the crystal lattice and experience resistance.[35–37] Analogously, a critical magnetic field disrupts the coordinated motion of electrons, causing trajectories to mismatch and thus resistance. In the case of the absence of electron vacancies in the chemical compound, the formation of "internal conductivity" becomes impossible, which excludes the possibility of realization of the superconducting state.[38] Therewith, it should be considered that superconductivity can arise due to various mechanisms depending on the nature of the material, and a universal model describing all cases does not exist.

For many chalcogenide glassy semiconductors, including metallic elements, in thin-film systems, the effect of rapid (~$10^{-10}$ s) switching from the high-resistance to low-resistance state under the influence of a strong electric field, accompanied by a jump-like increase in conductivity by several orders of magnitude, is observed.[39]

It is suggested that by controlling the width of the forbidden zone in superconductor or semiconductor materials, it is possible to create systems with controllable electronic transitions and implement controllable switches. Such an approach extends beyond the classical concepts of superconductivity, but it is alternative theoretical models that are of interest for explaining high-temperature superconductivity.

In confirmation of the need for a non-standard approach, the example of "atypical" superconductivity in the iron germanide $YFe_2Ge_2$ can be cited,[40] whose properties do not conform to the classical Bardeen-Cooper-Schrieffer (BCS) theory. By analyzing the electronic structure of $YFe_2Ge_2$, the presence of two electron vacancies was established, which corresponds to the observed critical temperature of about 10K (Fig. 1 – black circle[40]), demonstrating adequate agreement between theoretical calculations and experiment, despite the absence of cuprate or sulfide nature of the compound.

The results of model calculations and comparison of structural features show that an increase in the CuS content in sulfide compounds favors an increase in the number of electron vacancies, which raises the critical temperature of the superconducting transition (Fig. 1). The dependence is analogous to that observed in cuprates, where an increase in the number of CuO layers also leads to an increase in the superconductivity temperature.[41] Additionally, an increase in the proportion of trivalent chromium sulfide in the compound can also contribute to

the increase in the concentration of vacancies and, consequently, to the increase in the critical temperature, which confirms the significance of the number of electron vacancies as a key factor in the formation of high-temperature superconductivity. Also, chromium sulfide is hygroscopic, meaning it can absorb moisture from the air, which may affect the conductivity of the sample. The absorbed water could introduce additional charge carriers, such as protons or hydroxide ions, potentially increasing the conductivity. Additionally, moisture absorption might alter the electronic structure of the material, possibly modifying the valency of chromium ions or creating defects that affect conductivity. These changes could lead to variations in the material's superconducting properties, including the $T_c$. To minimize these effects, $Cr_2S_3$ should be handled and stored in a controlled, dry environment, and prior to measurements, the samples should be annealed under vacuum to remove any absorbed moisture and ensure more consistent and accurate conductivity data.

## 4. Conclusions

The study comprehensively evaluated superconducting properties of three-dimensional sulfide compounds containing transition metals, such as chromium and copper, with special emphasis on the influence of electronic vacancies in the crystal lattice. As a result of the synthesis and subsequent analysis of the structure, composition, and electrophysical properties of the films, the $Fe_2Cu_3S_6$ compound was found to possess superconductivity with a critical transition temperature of $T_c \approx 89$ K. The temperature transition was observed in a narrow temperature range, which may be related to ferromagnetic manifestations of residual iron, preventing superconductivity at lower temperatures. Based on elemental analysis and calculations, the study found that this compound contains about five electron vacancies, which helped to introduce the temperature coefficient of superconductivity for sulfides: $k_S = 17.8\ \mathrm{K}/Q_{elv}$. This coefficient was found to be greater than analogous values obtained for known oxide superconductors such as $La_2CuO_4$ and $Y_2Ba_4Cu_6O_{13}$ ($k_o \approx 11.6$–$11.7\ \mathrm{K}/Q_{elv}$), which may reflect a potential advantage of sulfide systems in the formation of high-temperature superconductivity.

The study also explored the concept of "internal electronic mobility" as a distinct chemical phenomenon, driven by the movement of electrons within the inner electron shells of transition metal atoms through localized electron vacancies. This model suggests that the presence of such vacancies facilitates the formation of unique electronic pathways within the crystal lattice, particularly in compounds with unstable or highly covalent structures, such as $Cr_2CuS_4$. Empirical observations indicate a linear correlation between the number of electron vacancies ($Q_{elv}$) and the temperature-dependent properties of ternary sulfide compounds. This relationship highlights the potential of Cr-Cu-S systems for developing materials with exceptional electronic characteristics, achievable under standard atmospheric conditions. Notably, the synthesis of these sulfide compounds is simpler and more cost-effective compared to the complex processes required for oxide or hydride materials. The trend observed in these systems underscores their promise for achieving advanced electronic properties, including high thermal stability and controlled conductivity, without the need for extreme synthesis conditions. This makes Cr-Cu-S compounds particularly attractive for applications requiring robust and tunable electronic behaviour.

Thus, within the framework of the study, the relation between the number of electron vacancies and the critical temperature of the superconducting transition was experimentally confirmed, which opens the way to the purposeful design of new classes of superconductors based on transition metals and sulfur. A limitation of this study is the lack of direct validation of the predicted models under high-pressure and magnetic field conditions. Prospects for research include the continued development of methods for the synthesis of sulfide compounds with an increased number of electron vacancies to achieve superconductivity at hotter temperatures.

## Conflict of Interest

The author declares no conflicts of interest.

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## ДОСЛІДЖЕННЯ НАДПРОВІДНИХ ВЛАСТИВОСТЕЙ СПОЛУК СУЛЬФІДІВ ПЕРЕХІДНИХ МЕТАЛІВ

***Анотація.*** *Метою цього дослідження було вивчення впливу електронних вакансій на надпровідні властивості сполук сульфідів перехідних металів (Cr, Cu) з метою оцінювання їхнього потенціалу щодо підвищення критичної температури переходу. Проаналізовано теоретичні моделі та експериментальні дані щодо надпровідності на основі сполук міді, хрому та сульфідів. Для аналізу використано дані синтезу та стабільності сполуки $FeCuS_2$, а також розраховані температурні коефіцієнти для різних надпровідних матеріалів, ураховуючи оксиди та сульфіди. У дослідженні розглянуто сполуку $Fe_2Cu_3S_6$, яка виявляла надпровідність за 89 K, з вузьким інтервалом переходу та ознаками феромагнітного впорядкування. Експериментальні дані свідчать, що вакансій електронів у кристалічній решітці впливають на провідність, а це сприяє виникненню надпровідного стану. Модельні розрахунки виявили лінійну залежність між кількістю вакансій електронів $Q_{elv}$ і температурою переходу $T_c$ у серії сульфідних систем, зокрема Cr-Cu-S. Це створює перспективи для пошуку надпровідників із високими температурами переходу. Порівняння з іншими матеріалами, такими як оксидні сполуки, показало, що сульфіди можуть виявляти вищі критичні температури надпровідності. Практичне значення дослідження полягає у розробленні нових підходів до проєктування сульфідних сполук, що сприяють виникненню надпровідного стану за кімнатних температур.*

***Ключові слова:*** *вакансії електронів, сульфідні сполуки, кристалічна решітка, халькогеніди.*