
Effect of metamaterial engineering on the superconductive properties of ultrathin layers of NbTiN

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Engineered superconductors: historic overview

- Proposals of 1D and 2D superconductors:

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Possibility of Synthesizing an Organic Superconductor*

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(Received 13 November 1963; revised manuscript received 27 January 1964)

V. L. Ginzburg, „On surface superconductivity“, *Phys. Lett.* 13, 101 (1964).

- Electromagnetic approach to superconductivity:

$$V(\vec{q}, \omega) = \frac{4\pi e^2}{q^2 \epsilon_{\text{eff}}(\vec{q}, \omega)}$$

ϵ should be negative and small!

D.A. Kirzhnits, E.G. Maksimov, D.I. Khomskii, “The description of superconductivity in terms of dielectric response function”, *J. Low Temp. Phys.* 10, 79 (1973).

New solution: artificial high polarizability metamaterials

I.I. Smolyaninov and V.N. Smolyaninova, “Metamaterial superconductors”, *Phys. Rev. B* 91, 094501

- Nanofabrication has reached a few nanometers scale – ability to engineer artificial material, which has enhanced superconducting critical temperature

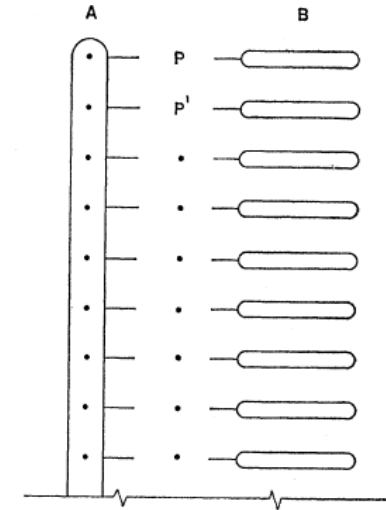
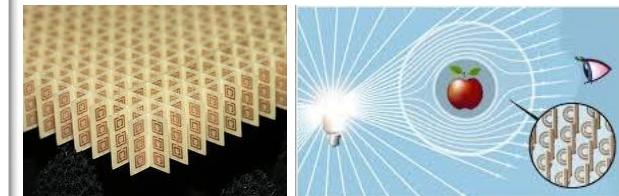


FIG. 1. Proposed model of a superconducting organic molecule. The molecule A is a long unsaturated polyene chain called the “spine.” The molecules B are side chains attached to the spine at points P, P', ...



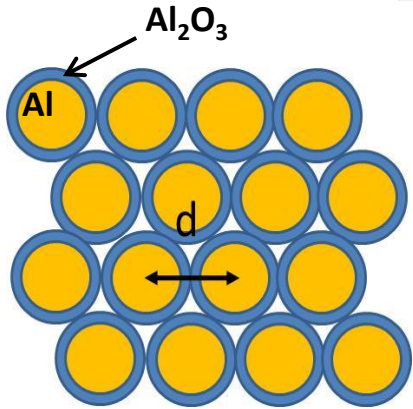
Recent developments in metamaterials

Artificial “metamaterials” may be created from much larger building blocks than atoms, and the electromagnetic properties of these fundamental building blocks (“meta-atoms”) may be engineered at will



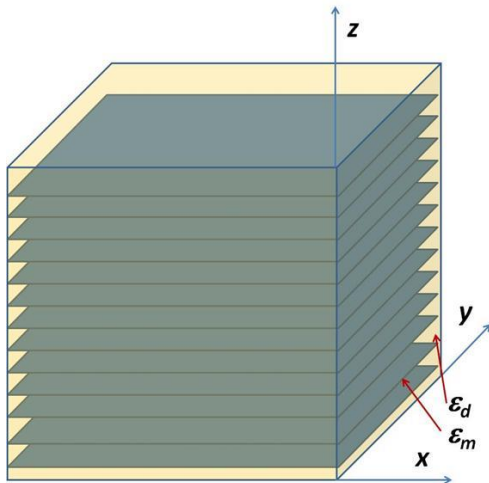
Engineering small and negative dielectric function $\epsilon(q,\omega)$

Counterintuitive approach:
adding insulator to superconductor to improve superconductivity



Epsilon near zero (ENZ) metamaterials: Al/Al₂O₃ core-shell metamaterials

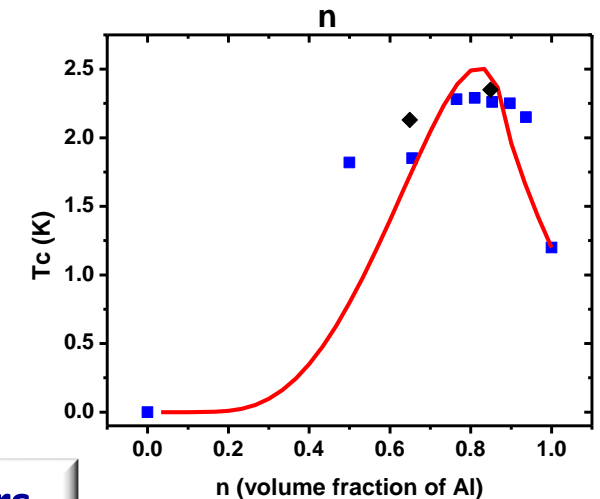
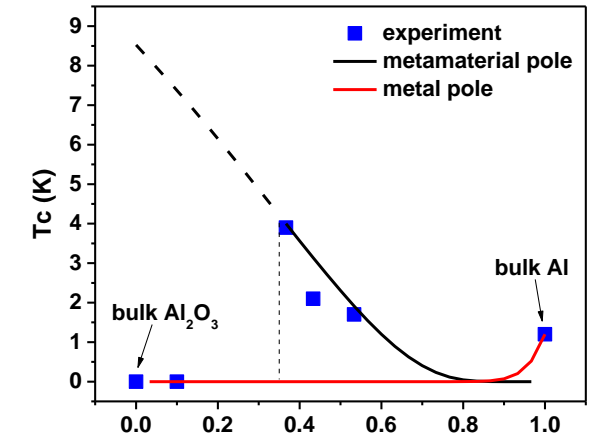
Scientific Reports 5,15777



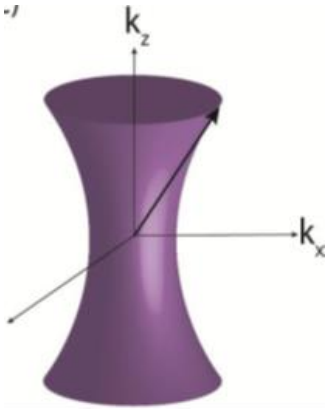
Hyperbolic metamaterials: Al/Al₂O₃ multilayers

Scientific Reports 6, 34140

Next step: test metamaterial approach on superconductors
with higher $T_c \rightarrow$ NbTiN hyperbolic metamaterials



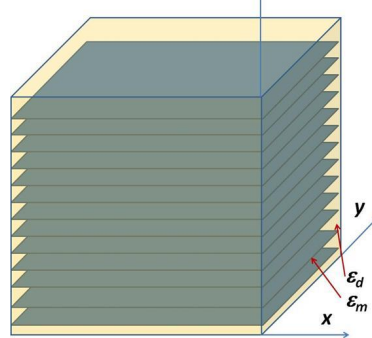
Hyperbolic metamaterial scenario



$$\frac{k_x^2 + k_y^2}{\epsilon_{zz}} + \frac{k_z^2}{\epsilon_{xx}} = \frac{\omega^2}{c^2}$$

$$\epsilon_{yy} = \epsilon_{xx} < 0; \epsilon_{zz} > 0$$

Hyperbolic metamaterial



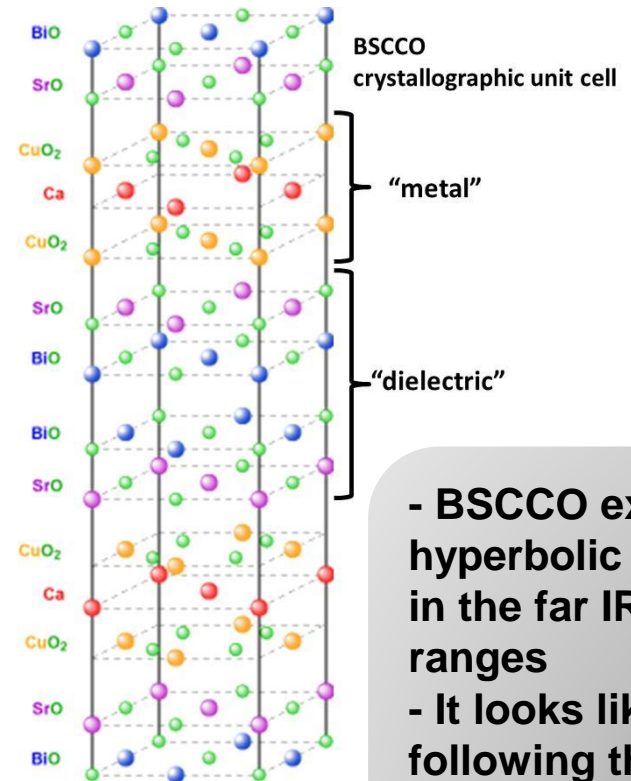
Effective Coulomb potential:

$$V(\vec{q}, \omega) = \frac{4\pi e^2}{q_z^2 \epsilon_{zz}(\vec{q}, \omega) + (q_x^2 + q_y^2) \epsilon_{xx}(\vec{q}, \omega)}$$

diverges at: $q_z^2 \epsilon_{zz}(\vec{q}, \omega) + (q_x^2 + q_y^2) \epsilon_{xx}(\vec{q}, \omega) \approx 0$

- Electron-electron interaction is strongly enhanced in hyperbolic metamaterials
- The best choice of geometry appears to be metal/dielectric layered structure

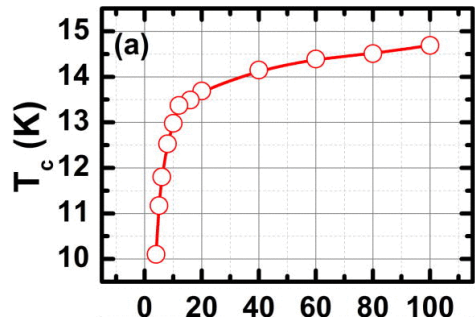
High T_c superconductor as hyperbolic metamaterial



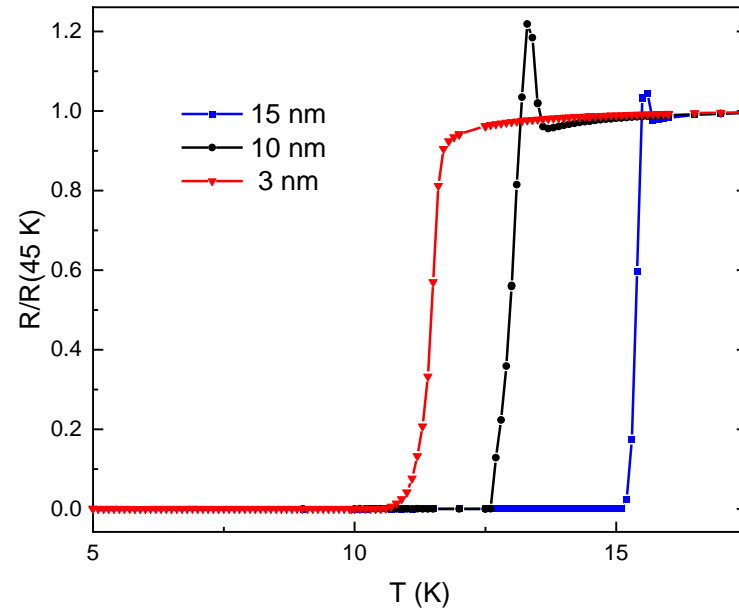
- BSCCO exhibits hyperbolic behaviour in the far IR and THz ranges
- It looks like nature is following the metamaterial recipe!

Effect of metamaterial engineering on the T_c and H_c of ultrathin layers of NbTiN

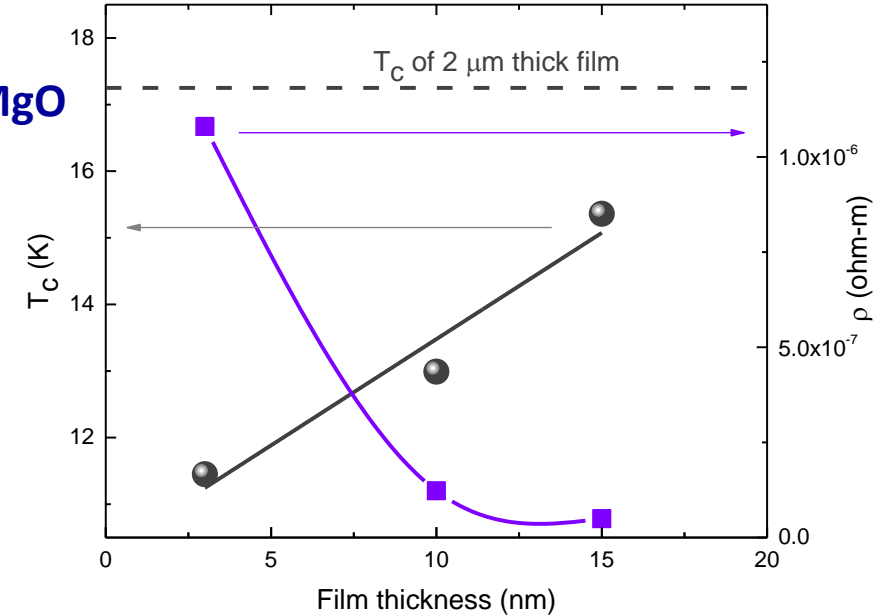
In ultrathin layers of NbTiN T_c depends on film thickness



Appl. Phys. Lett. 107, 122603



NbTiN on MgO



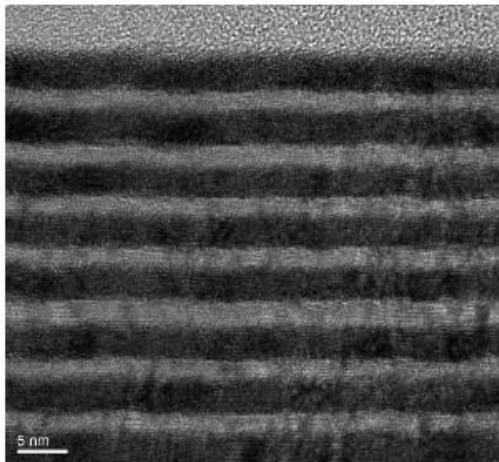
Apply metamaterial engineering to increase T_c

Samples

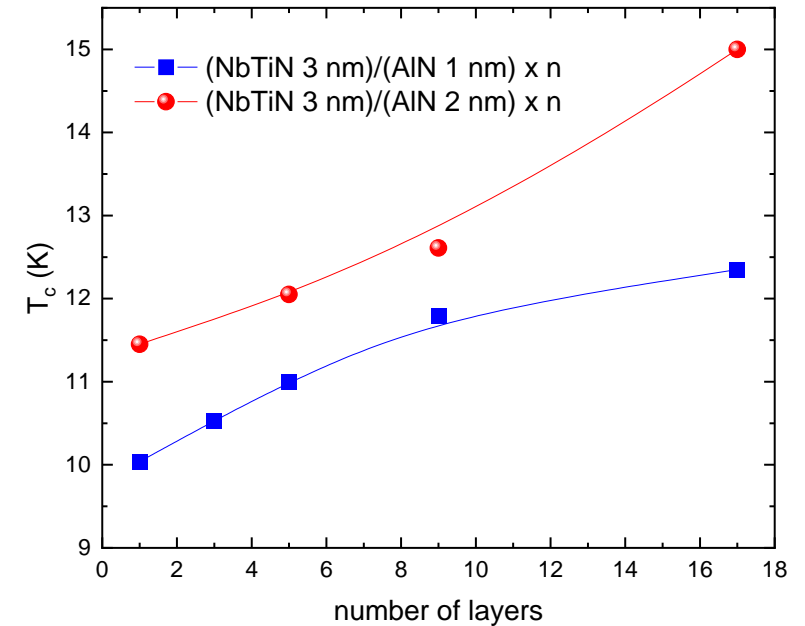
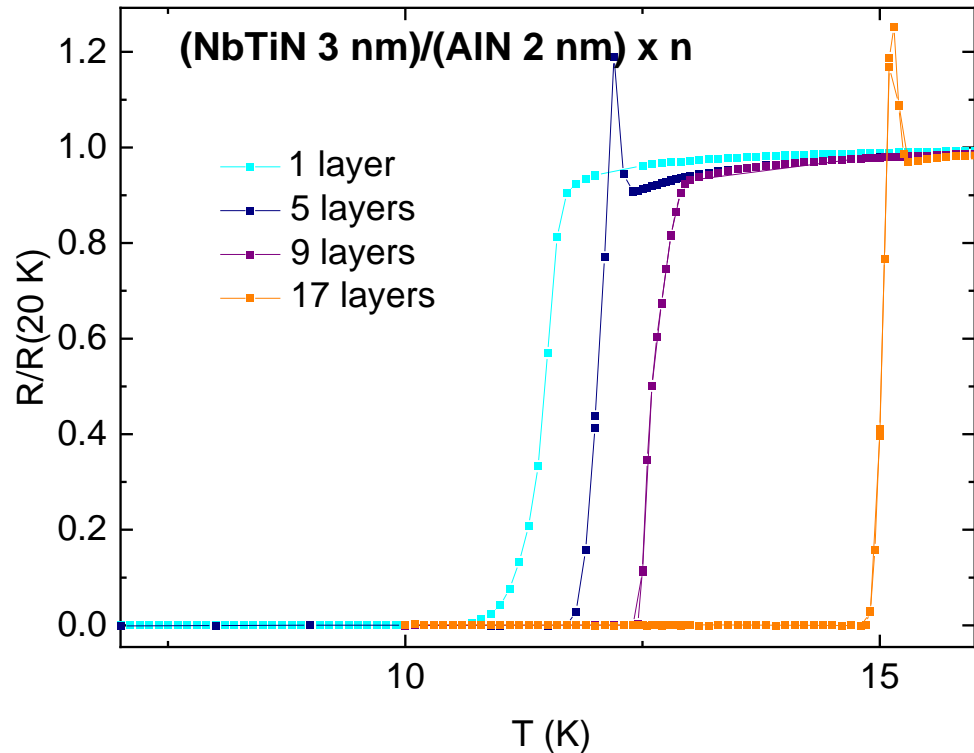
NbTiN films were grown using reactive high-power impulse magnetron sputtering (R-HiPIMS)

$$2 \text{ nm} < t_{\text{NbTiN}} < 3.6 \text{ nm}$$

$$0.8 \text{ nm} < t_{\text{AlN}} < 2 \text{ nm}$$



T_c dependence on number of layers



The T_c increases with the number of layers, with largest increase of the T_c by 3.6 K (by 32%) for 17 NbTiN/AlN layers, as expected for a superconducting hyperbolic metamaterial. Indeed, such a material with a large number of building NbTiN/AlN blocks can be considered to be a “metamaterial”.

Polarization reflectometry: anisotropy of the dielectric function

$$R_s = \left| \frac{\sin(\theta - \theta_{ts})}{\sin(\theta + \theta_{ts})} \right|^2$$

$$R_p = \left| \frac{\epsilon_1 \tan \theta_{tp} - \tan \theta}{\epsilon_1 \tan \theta_{tp} + \tan \theta} \right|^2$$

$$\theta_{ts} = \arcsin\left(\frac{\sin \theta}{\sqrt{\epsilon_1}}\right)$$

$$\theta_{tp} = \arctan \sqrt{\frac{\epsilon_2 \sin^2 \theta}{\epsilon_1 \epsilon_2 - \epsilon_1 \sin^2 \theta}}$$

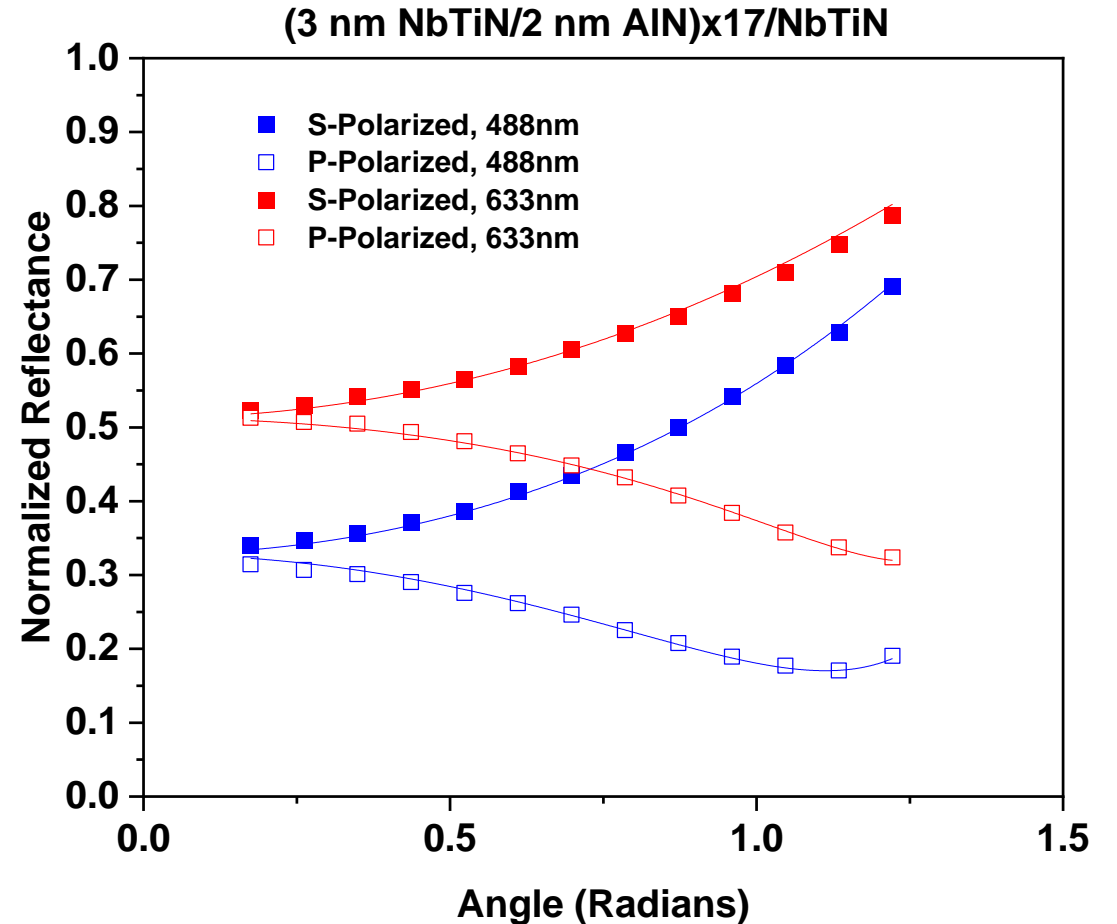


$$\epsilon_{\text{in plane}} = -3.8 + 7.2i$$

$$\epsilon_{\text{out of plane}} = 5.0$$

$$\epsilon_{\text{in plane}} = -0.51 + 4.7i$$

$$\epsilon_{\text{out of plane}} = 1.3 + 7i$$



Hyperbolic properties of multilayers have been achieved: $\epsilon_{\text{in plane}} < 0$; $\epsilon_{\text{out of plane}} > 0$

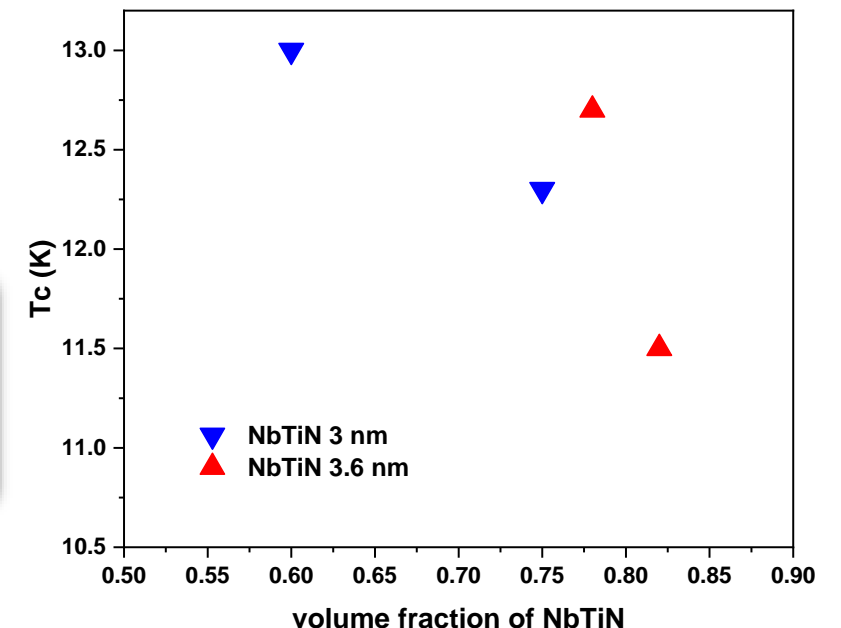
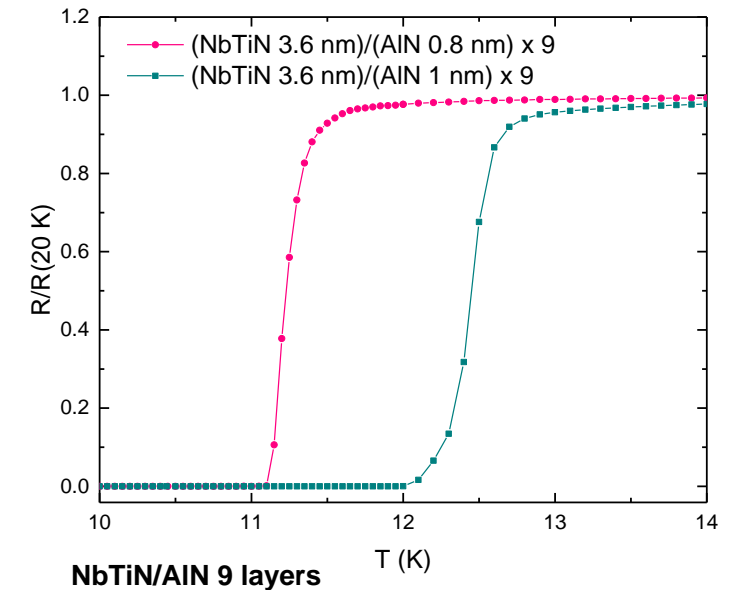
Variation of volume fraction of superconductor

The magnitude of the electron-electron interaction depends on an anisotropic dielectric function, which, in turn is a function of the volume fraction of the metal, n . In the case of a layered hyperbolic metamaterial, the dielectric function dependence on n can be found from the Maxwell-Garnett approximation:

$$\varepsilon_1 = n\varepsilon_m + (1-n)\varepsilon_d \quad \varepsilon_2 = \frac{\varepsilon_m \varepsilon_d}{(1-n)\varepsilon_m + n\varepsilon_d}$$

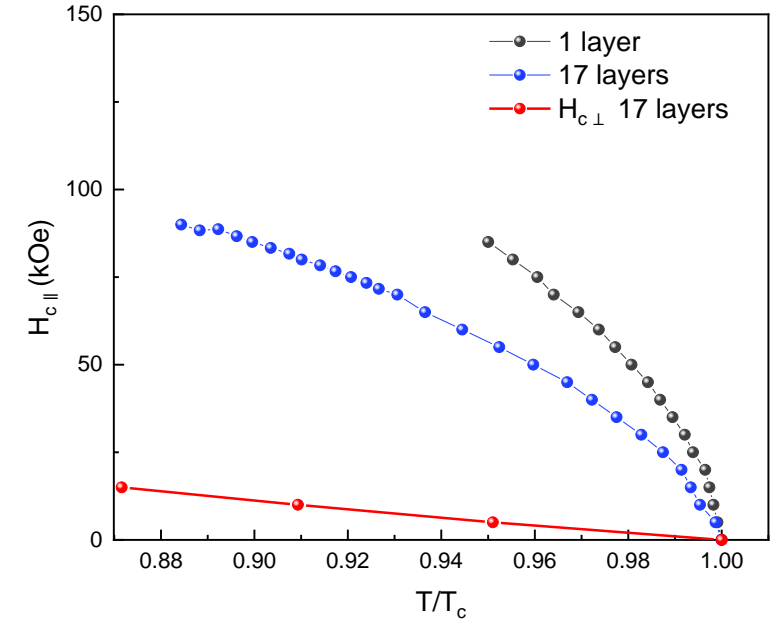
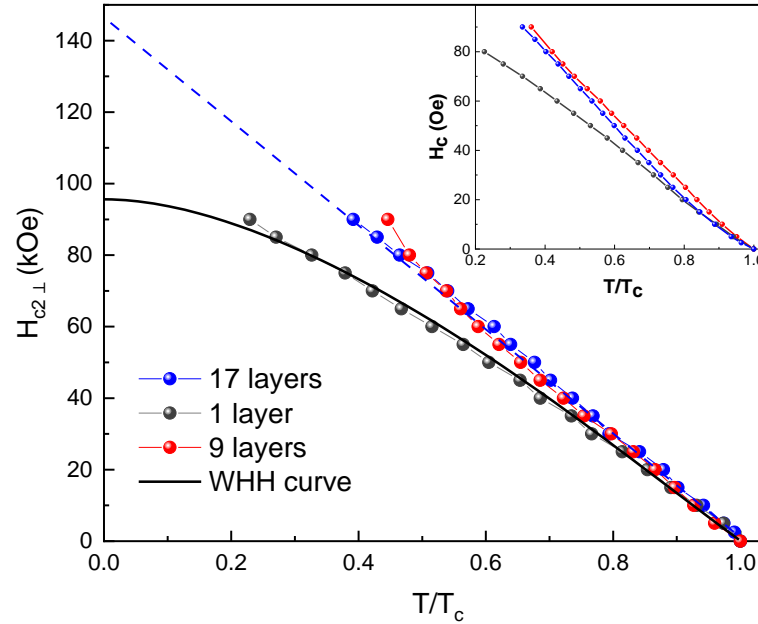
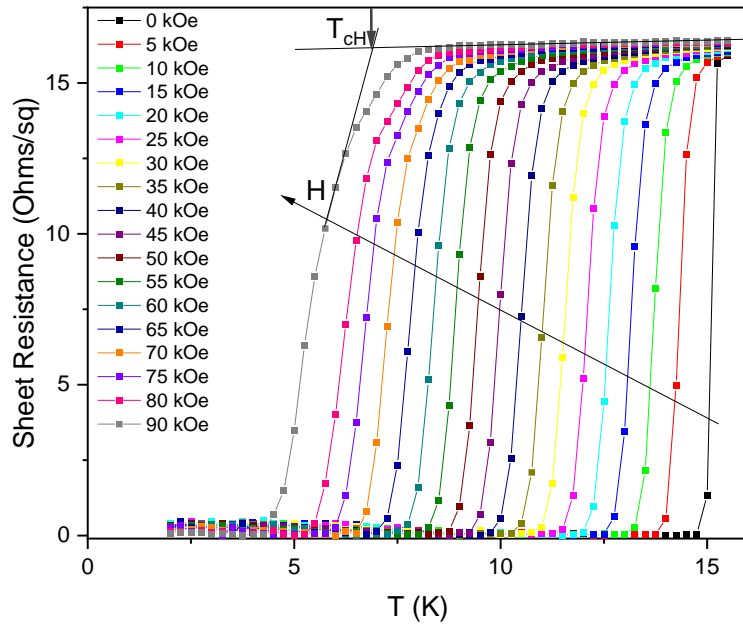
T_c should depend on the volume fraction of metal in the metamaterial. To test this assumption, the volume fraction of the dielectric in the multilayers was varied, while the thickness of the metal layer remained constant, since the T_c of a single layer depends on its thickness.

T_c of these superconducting hyperbolic metamaterials decreases with increasing volume fraction of the metal. The results are consistent with those for aluminium and tin based hyperbolic metamaterials.



Anomalous behavior of upper critical field in NbTiN-based multilayers

(NbTiN 3 nm)/(AlN 2nm) x n multilayers



- While $H_{c2\perp}$ of the single NbTiN layer follows the Werthamer–Hefand–Hohenberg (WHH) model, the multilayers exhibit a higher $H_{c2\perp}$ with an anomalous linear temperature dependence, or a slight positive curvature.
- Extrapolated $H_{c\parallel}(0) \sim 420 \text{ kOe}$

Ultrathin superconducting films are known to exhibit extremely high values of the critical magnetic field H_c . However, the T_c of a single layer of ultrathin films is significantly reduced.

We have demonstrated the ability to maintain the T_c of a superconducting coating while keeping (or even increasing) its critical magnetic field at the same level as for the ultrathin films. This result can lead to many advances in technological applications of superconductors.

Conclusions

NbTiN/AlN multilayered metamaterials with ultrathin layers

- exhibit up to a 32% enhancement of T_c with respect to the T_c of a single ultrathin NbTiN layer.
- This T_c increase can be attributed to an enhanced electron-electron interaction in superconducting hyperbolic metamaterials.
- The critical fields in these multilayers are high and have anomalous linear temperature dependence in the perpendicular to the magnetic field direction.

These results demonstrate that the metamaterial approach to superconductor engineering can enable the increase of H_{c2} as well as T_c .

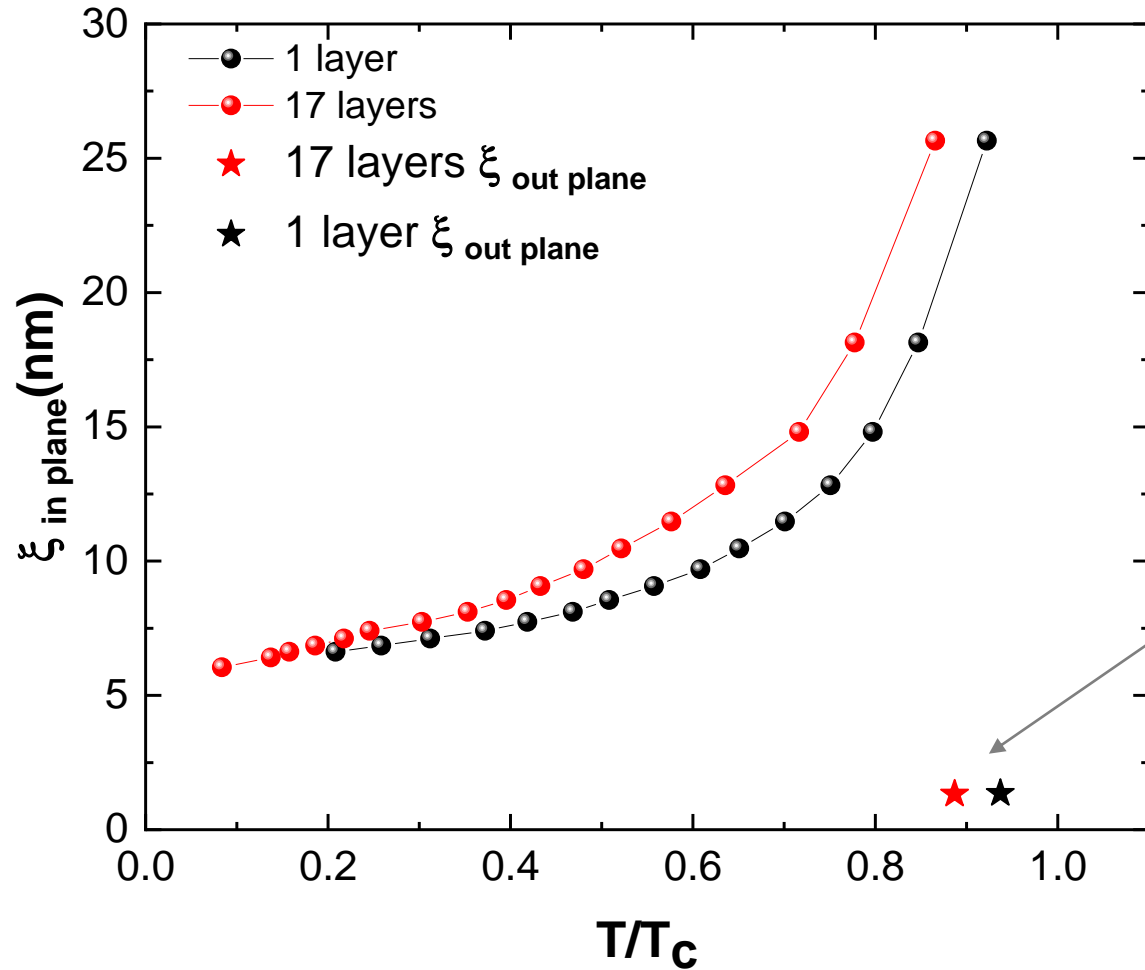


Anisotropy of coherence length

$$H_{c2\perp}(T) = \Phi_0 / 2\pi\xi_{\parallel}(T)^2$$

ξ_{\parallel} - in plane coherence length

(NbTiN 3 nm)/(AlN 2nm) x n multilayers



$$H_{c2\parallel}(T) = \Phi_0 / 2\pi\xi_{\parallel}(T)\xi_{\perp}(T)$$

ξ_{\perp} - out of plane coherence length

$$6 \text{ nm} < \xi_{\parallel} < 25 \text{ nm}$$

$$\xi_{\perp} \approx 1.4 \text{ nm}$$