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Supplementary information

Magnetic relaxation and three-dimensional critical fluctuations in B-doped Q-Carbon - A high-temperature superconductor

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Additional information:

Figure S1 depicts the results from high-resolution electron microscopy, electron energy loss spectroscopy (EELS) and unpolarized Raman spectroscopy of B-doped Q-carbon. The filamentary structure (as seen in the SEM figure S1(a)) is a result of interfacial instability during the super undercooling process when nanosecond ArF laser is used.[1] Previous reports indicate a negative electron potential (*w.r.t* diamond-like carbon) of Q-carbon structures which makes it glow with the application of a stage voltage bias.[2] As it is evident from the figure S1(a) that the formed B-doped Q-carbon filamentary structures are well connected which is a critical criterion for the electrical measurements. Figure S1(b) depicts unpolarized Raman spectroscopy of the as-deposited B-C layer and B-doped Q-carbon using 532 nm as the excitation source. As it is evident from the figure S1(b) there is a considerable increase in the sp^3 fraction in B-doped Q-carbon (~84%) as compared to as-deposited B-C sample (~70%). The inset of the figure S1(b) indicates the fitted peaks at 1105 cm^{-1}, 1322 cm^{-1}, and 1567 cm^{-1} of B-doped Q-carbon. The first two vibrational modes (lower wavenumbers) correspond to sp^3 hybridized carbon whereas the last peak corresponds to sp^2 hybridized carbon. Due to the ultrafast melting and subsequent quenching process in B-doped Q-carbon, there is an increase in the sp^3 fraction. This sp^3 fraction can also be controlled by the laser (wavelength, pulse duration) and substrate (thermal conductivity) parameters which results in the formation of 100% sp^3 crystallite diamond (small undercooling) or Q-carbon (large undercooling).[2] As mentioned before, B-doped sp^2 hybridized carbon (non-superconducting entities) which is present in the B-doped Q-carbon may act as effective pinning sites thereby increasing the values of pinning potential (near T_c). This results in superior superconducting properties of B-doped Q-carbon. Q-carbon consists of randomly arranged sp^3 tetrahedra with sp^2 bonding between them. Figure S1(c) depicts the cross-sectional

high angle annular dark field (HAADF) image of the formed superconducting B-doped Q-carbon on c-sapphire substrate, suggesting its uniform amorphous nature. Further details about the B-doping concentration and distribution in Q-carbon were obtained using electron energy-loss spectroscopy (EELS). Figure S1(d) shows a representative as-acquired and background subtracted EEL spectra of B-doped Q-carbon containing B-K (188 eV) and C-K edge (284 eV). Based on the EELS quantification,[3] B concentration is estimated to be 17.0±1.0 at% in Q-carbon. A uniform distribution of B in Q-carbon is also observed which has been reported earlier.[4] The spectrum contains the characteristic π* and σ* peaks associated with the K-edge of B and C, which indicates the presence of C and B bonded in both sp^3- and sp^2-hybridized state. From the analysis of C-K edge, sp^3 fraction of C in B-doped Q-carbon is estimated to be ~80% which is consistent with the Raman spectroscopy measurements. Additionally, the sp^3 fraction of B, as calculated from the K-edge of B, is determined to be 0.6 which resembles the substitutional B atom in diamond lattice and contributes to the superconducting nature of B-doped Q-carbon.

The Ginzburg-Landau coherence length (ε_L) can be calculated using the equation (1):

$$\varepsilon_L = \left[\Phi_0 / 2\pi H_{c2}(0)\right]^{0.5} \quad (1)$$

where, Φ_0 and $H_{c2}(0)$ denote the superconducting magnetic flux quantum and upper critical field at 0 K, respectively. Using the value of $H_{c2}(0)$ as 5.42 T, the Ginzburg-Landau coherence length can be calculated. The Ginzburg-Landau parameter (κ) is calculated using the equation (2):

$$\frac{H_{c1}(0)}{H_{c2}(0)} = \frac{ln(k)}{k^2 \times 2\sqrt{2}} \quad (2)$$

The value of $H_{c1}(0)$ is taken as 0.1 T. In the figure S2(a), we have plotted Difference *vs* κ, where $Difference = \frac{H_{c1}(0)}{H_{c2}(0)} - \frac{ln(k)}{k^2 \times 2\sqrt{2}}$. Therefore, we get the values of κ when the value of difference crosses the y=0 line. As it is evident from figure S2(a), the values of κ are 1.06 and 5.04. We

have reported the value of κ as 1.06 ($\kappa > 1/\sqrt{2}$) when the difference curve first crosses the y=0 line. This also indicates a type-II superconductivity in B-doped Q-carbon. The value of penetration depth (λ_d) can be calculated using the equation (3):

$$H_{c1}(0) = (\Phi_0/4\pi\lambda_L^2) ln (\lambda_L/\varepsilon_L) \quad (3)$$

In the attached figure S2(b), we have plotted Difference *vs* λ_L, where $Difference = H_{c1}(0) - (\Phi_0/4\pi\lambda_L^2) ln (\lambda_L/\varepsilon_L)$. Therefore, we get the values of λ_L when the value of difference crosses the y=0 line. As it is evident from figure S2(b), the values of λ_L are 8.4 and 57.4 nm. We have reported the value of λ_L as 8.4 nm when the difference curve first crosses the y=0 line. We have also plotted the Difference *vs* λ_L (in figure S2(c)) for Nb superconductor having the values of lower critical field, λ_L and ε_L as 198 mT, 58 nm and 38 nm. Figure S2(c) is in excellent accordance with the calculations mentioned above.

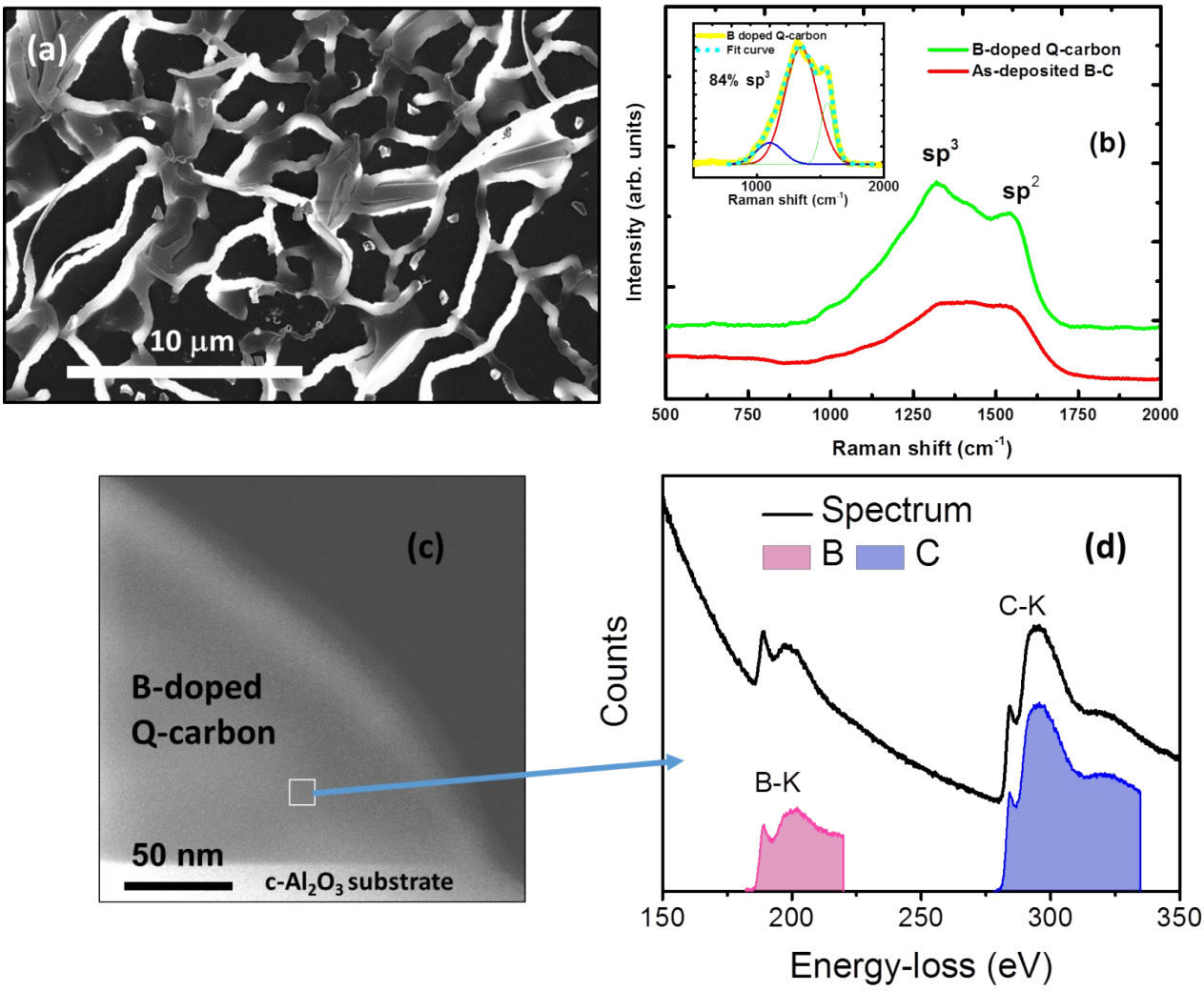

Figure S1: (a) High-resolution SEM showing connected fillamentary B-doped Q-carbon; (b) Unpolarized Raman spectroscopy of as-deposited B-C and B-doped Q-carbon thin film with the inset showing 84% sp^3 in the B-doped Q-carbon phase; (c) HAADF image of the formed superconducting B-doped Q-carbon on c-sapphire substrate; and (c) EELS of B-doped Q-carbon showing B-K and C-K edges.

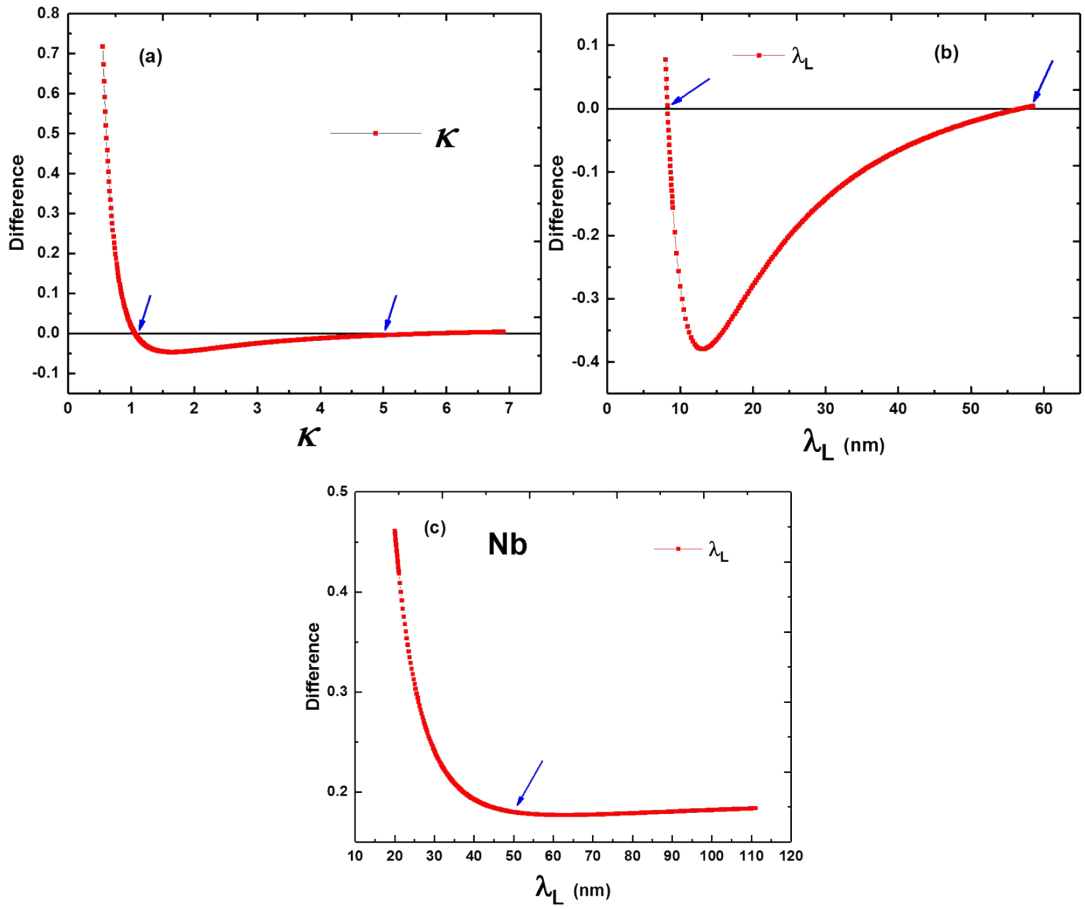

Figure S2: (a) The calculated values of κ , (b) λ_L in B-doped Q-carbon; and (c) Calculated value of λ_L in Nb.

References:

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