Dear Editor,

Please find our manuscript entitled “Effect of C-concentration on Fe self-diffusion and physical properties of $Fe_{1-x}C_x$ ($0 \le x \le 0.50$) thin films” by Prabhat Kumar, O. Leupold, I. Sergueev, H.-C. Wille and Mukul Gupta for the publication in *Journal of Physics: Condensed Matter*. We hereby confirm that this manuscript is not being considered for publication elsewhere.

In this work, we focused on $Fe_{1-x}C_x$ ($0.0 \le x \le 0.5$) thin films deposited by co-sputtering from Fe and C targets using direct current magnetron sputtering technique at the substrate temperature of 300 K. Generally, iron carbide (Fe-C) compounds possess positive enthalpy of formation, therefore, the formed structures are either nanocrystalline or amorphous at the ambient conditions. The magnetic and other properties of such systems can be tuned by tuning of grain size of magnetic species with the carbon concentration which makes them useful in various magnetic devices. In addition, in recent years, it has also gained attention due to the higher specific capacity (Electrochimica Acta 170 (2015) 16–2) for anode materials in lithium-ion batteries. In this viewpoint, the structure should be known which is not well understood hitherto. Previously, the structures of $Fe_{1-x}C_x$ thin films deposited by sputtering from a compound [Fe + C] targets were investigated using Mössbauer spectroscopy, and usually, it is found to be the growth of a magnetic structure. Furlan et al., [J. Phys.: Condens. Matter 27 (2015) 045002] investigated structures of amorphous $Fe_{1-x}C_x$ ($0.21 \le x \le 0.74$) thin films deposited from compound targets also, and observed the presence of iron-rich carbide phase and amorphous carbon-rich matrix. In addition, they also predicted the presence of a third carbide phase in terms of the charge transfer effect observed from x-ray absorption spectroscopy. Further, Zhao et al., [Electrochimica Acta 170 (2015) 16–2] observed two Fe-C structures in $Fe_{1-x}C_x$ ($0.47 \le x \le 0.97$) thin films deposited by simultaneous sputtering (co-sputtering) from individual Fe and C targets.

In this work, we utilized co-sputtering to prepare $Fe_{1-x}C_x$ thin films which is a rarely used method; however, it offers uniform mixing of species compared to that from the compound targets [Phys. Rev. Materials 4, 013402 (2020)]. The nobility of our work is to study the Fe self-diffusion that is taking place during the growth itself and then by subsequently measuring the magnetization of a thin (3 nm) $^{57}Fe_{1-x}C_x$ layer sandwiched between thick natural $Fe_{1-x}C_x$ layers using nuclear resonant scattering (NRS) – a powerful synchrotron-based technique to determine the magnetic structure, this technique is based on the basic principle of Mössbauer effect. The NRS measurement reveals the formation of one to three Fe-C structures with increasing carbon concentrations. Meanwhile, we found that during the growth of structures Fe self-diffusion was increased appreciably at the lower carbon concentrations compared to that in pure Fe film. However, it decreases above a carbon concentration systematically.

By suitably using complementary characterization techniques like x-ray diffraction (XRD), atomic force microscopy (AFM), x-ray absorption near-edge spectroscopy (XANES), SQUID vibrating sample magnetometer (SQUID-VSM), magneto-optic Kerr effect (MOKE) and grazing incidence NRS, detailed information about the crystal structure, surface morphology, local electronic and magnetic structure has been obtained. These measurements suggest that the Fe-C structures are arranged in a self-organized multilayered or cluster structures. The formation of such structures has an interesting impact on the magnetic properties in terms of magnetic anisotropy which increases with carbon concentrations. Our observation shows that by suitably tuning the growth conditions through the control of the diffusion process such magnetic properties can be further enhanced. Thus, the outcome of the present work is unique, and certainly will be helpful to the scientific community in the understanding of the structural and magnetic properties of amorphous iron carbide systems. Therefore, we kindly request you to consider this manuscript for publication in the *Journal of Physics: Condensed Matter*.

Yours Sincerely,

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