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Correlation of microstructures and exsolution lamellae in natural pyrrhotites and magnetic properties

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Pyrrhotite is a magnetic iron sulfide of the general non-stoichiometric composition Fe_{1-x} S ($x < 0.125$) and occurs widespread in Earth's crust, where it can contribute significantly to the magnetization of rocks. Pyrrhotite stands out due to its highly variable, and composition dependent magnetic properties. At ambient temperature two principle pyrrhotite types can be distinguished: 4C-pyrrhotite ($Fe_{0.875}S$, monoclinic pyrrhotite) and NC-pyrrhotite ($Fe_{0.88-0.92}S$, 'hexagonal' pyrrhotite). The 4C variant is ferrimagnetic, while the NC variants appear antiferromagnetic at room temperature. 4C-pyrrhotite has a well defined composition and crystal structure, but NC-pyrrhotites strongly vary and show complex crystallographic superstructures that arise from the ordering of vacancies on Fe lattice sites. In both varieties the ordering of Fe vacancies determines the magnetic properties, but exact mechanisms, especially in case of NC-pyrrhotites, are not well constrained. Little is known about the nature of several magnetic and structural transitions at low (≈ 30 K), and elevated ($470-540$ K) temperatures (NA- and MC-pyrrhotites). A major complication in the study of pyrrhotites arises from exsolution lamellae that involve different pyrrhotite superstructures and other intergrown phases (e.g. magnetite). In order to elucidate how the intergrowths of different pyrrhotites and other magnetic minerals affect the bulk magnetic properties, we have performed low- and high-temperature magnetic studies on a suite of natural samples that have been extensively characterized by electron microprobe analysis transmission electron microscopy, electron backscatter diffraction, and X-ray diffraction. The set includes pyrrhotite from Drag at the Tysfjord, Norway (TYS, NC plus troilite exsolution lamellae), Bodenmais, Germany (BOD, nearly pure NC with subordinate 4C and magnetite exsolution), Nyseter Mine near Grua, Norway (NYS, NC with 4C exsolution), Sta. Eulalia, Mexico (EUL, NC with 4C exsolution), and Dalnegorsk, Russia (DAL, 4C with subordinate NC exsolution). Zero-field cooling and field cooling warming curves of all samples show the low-temperature transition of pyrrhotite, close to 35 K, but some also show a transition near 15 K. For samples from BOD and TYS, the two cooling procedures lead to different warming curves. The high-temperature NC-NA transition close to 493K occurs in all samples, but with widely varying intensity. The NC-NA transition is clearly visible in BOD, TYS, NYS. In EUL it only leads to a small increase in saturation magnetization, while in DAL it is almost only recognizable in the low-field susceptibility. The $M_s(T)$-curves fade out at ≈ 600 K without a clear transition, but all $M_{rs}(T)$-curves show a clear zero transition between 580 K and 590 K. The data suggest that in addition to the relative proportion of 4C- and NC-pyrrhotite, the magnetic properties are influenced by subtle differences in the total NC superstructure and/or the NC/4C interface.