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Magnetic correlations in the disordered ferromagnetic alloy Ni-V revealed with small angle neutron scattering

We present small angle neutron scattering (SANS) data collected on polycrystalline Ni$_1-x$V$_x$ samples with $x \geq 0.10$ with confirmed random atomic distribution. We aim to determine the relevant length scales of magnetic correlations in ferromagnetic samples with low critical temperatures $T_C$ that show signs of magnetic inhomogeneities in magnetization and $\mu$SR data. The SANS study reveals signatures of long-range order and coexistence of short-range magnetic correlations in this randomly disordered ferromagnetic alloy. We show the advantages of a polarization analysis in identifying the main magnetic contributions from the dominating nuclear scattering.

Formation of ferromagnetism in metals is still an active field for discovery of novel phases and mechanisms in condensed matter physics. In particular, the control of disorder and determination of how inhomogeneities affect magnetic properties remains a significant challenge. Small angle neutron scattering (SANS) is one of the prime methods to characterize magnetic material at the nanoscale. It has revealed important insight in the complex structure formation of inhomogeneous magnets with defects or internal structures from bulk alloys to amorphous and nanocrystalline magnetic materials. In this study we focus on the binary transition metal alloy Ni$_1-x$V$_x$, that presents an example of a diluted inhomogeneous ferromagnet produced by random atomic distribution. The onset of ferromagnetic order of Ni at $T_C = 630$ K is suppressed towards zero with sufficient V concentration of $x_C = 0.116$. Previous magnetization and $\mu$SR studies show signatures of fluctuating clusters from Ni-rich regions for paramagnetic samples with $x > x_C$. These persist also into the ferromagnetic state close to $x_C$ and coexist with the static order evolving below $T_C$. With SANS we aim to measure the magnetic cluster sizes and their effect on the static order in this random disordered system. We present a SANS study with polarization analysis to extract magnetic scattering that would otherwise be dominated by nuclear scattering.

For this study we used the same polycrystalline samples of Ni$_1-x$V$_x$ that were prepared for optimal random distribution and characterized by several methods from previous studies. Several pellets of 3 mm diameter of each concentration were wrapped in Al foil and mounted on Al sample holder framed with Cd-mask and connected to the cold plate of the cryostat. The SANS experiments were performed at GPSANS, HFIR, Oak Ridge National Lab and at NG7SANS, NCNR, NIST. We show detailed data from NIST of Ni$_{0.90}$V$_{0.10}$ samples using also polarized neutrons (tracking the polarization (p) state before and after sample). The SANS intensity was collected in the $xy$-plane on a 2D detector at different distances to cover a wave vector $(Q)$-range of $(0.06-1)$ nm$^{-1}$ with neutron wavelengths of 0.55 nm and 0.75 nm. Taking advantage of supermirror polarizer and He-cell as spin analyzer as described in detail before, we collected separately non spin flip (NSF) scattering with unchanged p-state of the neutrons (DD and UU) and spin flip (SF) scattering with reversed p-state (DU and UD) from the sample. U, D refers to the neutron spins aligned UP, DOWN with respect to the...
neutron polarization axis defined by the external magnetic field. The magnetic field was applied in the x-direction ($B_{\text{max}} = 1.5$ T, $B_{\text{min}} = 7$ mT) perpendicular to the beam ($\perp z$), $\theta$ indicates the azimuthal angle within the $xy$-plane, with $\theta = 0^\circ$ in the horizontal $x$-direction. Horizontal or vertical data averages included a symmetric cone of width $\pm 30^\circ$ around $\theta = 0^\circ$ and $180^\circ$ or $\theta = \pm 90^\circ$ presented as SFH or SFV, respectively. Besides fully polarization-analyzed PASANS we collected also unpolarized or “non pol” data (NP) without tracking the p-state and “half pol” (HP) data without distinguishing the p-state after the sample (D and U) without a $^3$He-cell in the beam. The PASANS data were polarization corrected and reduced with the IGOR software.

First SANS studies of Ni$_{1-x}$V$_{x}$ could not resolve the weak magnetic scattering for paramagnetic samples with $x = 0.12$ but recent SANS data collected at NG7SANS, NIST and GPSANS, ORNL revealed a clear temperature-dependent signal in weak ferromagnetic samples with $x \leq 0.11$ that demonstrated that magnetic scattering can be resolved for ferromagnetic samples with a reduced average moment per Ni of $\mu = 0.03 \mu_{B}$. We show here the SANS data for $x = 0.10$ with $T_{c} \approx 50$ K collected at NIST to compare best full pol and non pol data. Fig. 1(a) presents the non polarized neutron scattering intensity as a function of the magnitude of the wavevector $Q$ collected as horizontal average NPH. NPH collects only transverse magnetic contributions $M_{x}^{2}$ and $M_{y}^{2}$ (not the longitudinal magnetic components $M_{z}^{2}$ along the field direction) beside the dominating nuclear contribution $N^{2}$ and other backgrounds $B_{\text{NP}}$.

$$NPH = NP(\theta = 0^\circ) = N^{2} + M_{x}^{2} + M_{y}^{2} + B_{\text{NP}} \quad (1)$$

The “non-magnetic” contributions are estimated by NPH collected in high fields ($NPH(B = 1.5$ T, $T < T_{c}$)) where aligned magnetic moments are expected to contribute only to the magnetic scattering in field direction $M_{z}^{2}$ not to the selected transverse components. The NPH difference of the total non pol scattering collected at different temperatures in low fields ($B = 7$ mT) and the high field data should finally reveal the magnetic scattering ($M_{x}^{2} + M_{y}^{2}$) as shown in Fig. 1(a).

Most magnetic scattering is found close to $T_{c} \approx 50$ K in the higher $Q$ range ($0.2$ nm$^{-1}$ - $1$ nm$^{-1}$). It can be approximated by a Lorentzian as expected for paramagnetic critical scattering of the Ornstein Zernike form with a correlation length $1/k$ increasing towards $T_{c}$. Below $T_{c}$ the magnetic intensity is significantly reduced in the higher $Q$ regime, and in addition, an increase of intensity at low $Q$ is noticed that follows a $1/Q^{4}$ dependence without any sign of saturation.

$$I_{\text{mag}}(Q) = \frac{D}{Q^{4}} + \frac{L}{(k^{2} + Q^{2})^{4}} \times F(Q) \quad (2)$$

The fit can be improved somewhat towards higher $Q$ by a form factor$^{1}$ $F(Q) = \exp(-\frac{1}{3}r_{0}^{2}Q^{2})$ with radius $r_{0} \approx 1$ nm that could indicate non-uniform magnetic scattering centers or be an artifact of non ideal background subtraction. Different than in homogeneous systems with a narrow critical regime with diverging correlations at $T_{c}$ typically observed with SANS$^{15}$ we find reduced length scales as seen in inhomogeneous ferromagnets e.g. in diluted ferromagnetic alloys$^{16}$ or diluted manganites. The correlation length of the visible magnetic contribution remains finite at $T_{c}$ ($1/k \approx 5$ nm) and even seems to grow further below $T_{c}$ at 30 K. At very low $T$ $T = 3$ K we still recognize similar transverse magnetic contributions as observed for $T_{c}$ with reduced amplitude but similar correlation length. This indicates that short-range fluctuations of the paramagnetic state are still left in the ferromagnetic state at low temperatures. The low-$Q$ upturn in the non pol data (NPH) is most likely due to the contrast of misaligned magnetic regions (domains) of a large scale ($>100$ nm) expected in a soft magnet of finite size at small fields. This domain term becomes apparent below $T_{c}$ indicating the onset of long-range order. However, it is difficult to extract the magnetic response from the huge nuclear $1/Q^{4}$ term due to grain boundaries in these polycrystalline samples.

Encouraged by these promising findings of sufficient magnetic scattering in the larger $Q$ regime, but uncertain about reasonable background estimates, we collected full polarized SANS. The clear advantage is the collection of pure spin flip (SF) data (DU+UD) recognizing electronic magnetic scattering through the angle $\theta$ dependence at constant Q.

$$SF(\theta) = M_{z}^{2} + M_{x}^{2}\cos^{2}\theta + M_{y}^{2}\cos^{2}\theta\sin^{2}\theta + B_{\text{SF}} \quad (3)$$

We noticed signs of anisotropy only in field direction $M_{x} > 0$ (see below) and did not consider transverse terms, $M_{y} = M_{z} = 0$ simplifies the spin flip intensity $SF(\theta)$ in Eq. (3). Fig. 2 presents the angle dependence of the SF data collected for a medium $Q$ range, (0.2 - 0.5) nm$^{-1}$, for Ni$_{10.90}$V$_{0.10}$ after a constant background has been subtracted. The solid lines represent fits using Eq. (3) that yield $M_{z}^{2}$ but also $M_{x}^{2}$ with less precision as presented in Fig. 3(a).

At high temperatures and very small magnetic fields SF($\theta$) follows a
pure \cos^2\theta dependence with \( M^2_y = M^2_x \) expected for isotropic paramagnetic fluctuations. The data confirm also that \( M^2_z = M^2_y \). At low \( T = 3\) K the SF data are shown for 50 mT and higher fields. At smaller fields the ferromagnetic sample (below \( T_c \)) depolarizes the neutron beam that PASANS cannot be analyzed. The magnetic signal \( M^2_y \) at 3 K in 50 mT is reduced to about 10% of \( M^2_y \) at \( T_c \). \( M^2_x \) is still similar to \( M^2_y \) in small fields. In higher fields \( M^2_y \) gets suppressed to reach very small values for 1.5 T. The confirmed isotropy underlines the fact that these short-range correlations stem from dynamic fluctuations that cold neutrons can collect up to the order of THz especially on smaller ranges. These SF data demonstrate that indeed a small fraction of magnetic fluctuations remain at low temperatures, that get further suppressed in higher magnetic fields.

Fig. 1(b) shows the \( Q \) dependence of the SF contrast, SFH-SFV, that evaluates \( M^2_y \). Since the magnetic response is isotropic for low fields this signal represents (1/3 of) the total magnetic fluctuations \( M^2_{\text{tot}} \). In this restricted \( Q \) regime a Lorentzian describes the data well. A finite \( r_0 \approx 1\) nm also improves the fit somewhat for large \( Q \). We did not aim to get more detailed nanostructures from these data using alternative descriptions including cluster distributions. The Lorentzian fit produces a correlation lengths for 47 K in the order of 7 nm that is similar at \( T = 3\) K in 50 mT. The estimate of \( 1/\kappa \approx (10 \pm 4)\) nm includes uncertainties caused by background variations contaminating the small magnetic signal that is resolved in a limited \( Q \) regime. Comparing panel (a) and (b) in Fig. 1 we see that the SF data confirm the non polarized magnetic estimates of fluctuations with similar magnitude (\( M^2_y \)) and length scales for common data sets in the higher \( Q \) regime. But below \( Q \approx 0.1\) nm the smaller SF data are difficult to resolve from dominating NSF data after polarization corrections and do not reveal the signatures of long-range order. If the 1/\( Q^4 \) upturn in the NPH difference is real magnetic scattering or an artifact of a nuclear origin or multiple scattering cannot be resolved with SF scattering and needs a different approach.
We cannot use the total non spin flip data, NSF, (DD+UU) to reveal the longitudinal magnetic component $M_z$ from the angular dependence since the nuclear scattering is dominating the signal. But we can take advantage of the difference response “DIF” between the two initial polarization direction (without registering spin flip), the NSF asymmetry or flipper difference from full pol data (DD-UU) and HP data (D-U) that yields an interference term of nuclear and magnetic origin. It signals a weak contribution from a center with a net magnetic component along the x-direction $M_x > 0$ in the presence of a strong nuclear contribution from the same center.

\[ DIF(\theta) = 2NM_x \sin^2 \theta \]  

(4)

DIF(\theta) is shown in the inset of Fig. 4 presenting the two maxima at $DIFV = 2NM_x$ according to Eq. (4). Even in the low $Q$ regime this structure can be resolved at low $T = 3 \text{ K}$ in sufficient high fields ($B \geq 50 \text{ mT}$). As shown in the main panel $DIF(Q) = 2NM_x(Q)$ can be presented by a $1/Q^2$ term and a small constant following Eq. (2) with a large parameter $x$. Since such $Q$ dependence is expected for nuclear scattering $N^2$ dominated by large grain boundaries we conclude a similar $Q$ dependence for $(M_x)^2$. Potential deviations of the form $1/(Q^2 + g^2)$ yield magnetic domain sizes larger than $1/\kappa = 50 \text{ nm}$ and the estimates of $2NM_x$ collected at different temperatures from the angle dependence $(A)$ and the $Q$ dependence $(D/Q_m)$ at low $Q = 0.1 \text{ nm}^{-1}$, $2NM_x \neq 0$ for $T < T_c$ while $2NM_x = 0$ for $T > T_c$. This interference term DIFV succeeds to resolve magnetic scattering expected for aligned ferromagnetic magnetic domains that form below $T_c$. The $1/Q^2$ dependence of $(M_x)^2$ reveals long-range magnetic domains. Simple estimates from neutron depolarization yield domain scales of micrometers at $T = 3 \text{ K}$. We expect that in a ferromagnet below $T_c$ fluctuations turn into long-range order, but in this inhomogeneous compound short-range fluctuations are more dominant than in defect-free, homogeneous systems. On one hand random defects produce distinct short-range correlations that are noticed at $T_c$ and are still present at low temperatures but on the other hand they do not destroy long-range order in this alloy.

Collecting SANS data with and without polarization analysis we gained new insight in the inhomogeneous ferromagnetic state of Ni$_{1-x}$V$_x$ with low critical temperatures $T_c$ below 50 K. In this paper we focus on Ni$_{0.90}$V$_{0.10}$. We found clear evidence of magnetic fluctuations in the larger $Q$ regime from spin flip (SF) contrast. From the fluctuations at $T_c$ a fraction of 10% remains at the lowest temperature of $T = 3 \text{ K}$ with similar correlation lengths of about 10 nm. In addition, the non spin flip (NSF) asymmetry (from full pol and half pol data) reveals large scale aligned magnetic domains in the lower $Q$ regime at low temperatures $T = 3 \text{ K}$ below $T_c$. Although these random defects cause short-range magnetic fluctuating clusters, long-range order still develops in this alloys. Similar features can be observed in Ni$_{0.78}$V$_{0.22}$ samples with $x = 0.11$ with smaller $T_c \approx 7 \text{ K}$. The challenge to resolve the smaller magnetic contribution from the overwhelming nuclear background is increased, but magnetic fluctuations remain and indication of aligned domains are present for low temperatures below $T_c$. More details will be presented elsewhere. We demonstrated that PASANS is a helpful method to clarify signatures of random dilution in alloys presenting magnetic correlations that persist in a wide range of length scales at low temperatures.

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