Europhysics Letters

OFFPRINT

Vol. 63 • Number 1 • pp. 56–62

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Published under the scientific responsability of the
European Physical Society
Incorporating
Journal de Physique Lettres • Lettere al Nuovo Cimento

EUROPHYSICS LETTERS 1 July 2003

Europhys. Lett., **63** (1), pp. 56–62 (2003)

Observation of spin-lattice coupling in the critical region of $Fe_{65}Ni_{35}$

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(received 4 July 2002; accepted in final form 8 May 2003)

PACS. 61.12.Ex – Neutron scattering techniques (including small-angle scattering).
PACS. 75.40.-s – Critical-point effects, specific heats, short-range order.
PACS. 75.50.Bb – Fe and its alloys.

Abstract. – The nuclear-magnetic interference in the critical small-angle scattering of polarized neutrons from invar $Fe_{65}Ni_{35}$ near $T_C = 485$ K was investigated. The interference caused by the spin-lattice coupling revealed the correlation between critical fluctuations of magnetization and fluctuations of the nuclear density. The magnetic scattering allowed one to unambiguously identify a shape of the scattering objects as one of the critical fluctuation. The q-dependence of the interference scattering revealed that the shape of the scattering nuclear objects is essentially the same as the magnetic one. Therefore, the shape and lifetime of the scattering nuclear object are correlated with and terminated by that of the magnetic fluctuation. It was found that the expansion of the lattice is $(\delta a/a) \approx 0.004$. The range of this expansion is of order 50 Å and depends on the temperature and magnetic field.

Introduction. – The classical invar has got its name after the discovery of its anomalously small value of the thermal expansion coefficient in a wide temperature range. Unusual behaviour of the lattice constant appears near the Curie temperature T_C. The invar effect is well understood as a volume increase associated with an increase of the magnetization or spin alignment as the temperature decreases below T_C. In other words, the effect is mainly explained as follows: as the sample is cooled, the increasing alignment of the spins causes the Fe-Fe repulsion, which creates an internal pressure and expands the lattice (see [1] and references therein). This interpretation stems from Weiss hypothesis [2] where he postulated the coexistence of two spin states for Fe atoms in FCC structure of the invar alloy: a high spin state (large moment, large volume) and a low spin state (small moment, small volume). Since that time numerous experimental and theoretical works have been published which support Weiss explanation of the invar effect. Among recent publications see, for example, refs. [3,4]. It was claimed recently that the invar effect may be explained if one takes into account non-collinear ordering of spins, rather than a change of the atomic magnetic moment [5]. The polarised neutron scattering experiment, performed short after, does not support the hypothesis of non-collinear state and proves that $Fe_{65}Ni_{35}$ is a collinear ferromagnet [6]. There was also a recent

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attempt to directly observe the transition from high-spin to low-spin state with increase of temperature using polarised neutron diffraction [7]. The results of the experiment show no change of moment in the temperature range between 100 K and 600 K in contradiction to the "two states" hypothesis. Thus, the question on the nature of the invar phenomena remains open. However, even less is known about the magnetic phase transition, which is the system undergoing it and how it relates with the appearance of the invar behaviour below T_C. The lack of knowledge on this matter is explained by a complexity of the phase transition, which, in turn, is caused by the non-trivial coupling between spin and lattice subsystems. Nevertheless, due to the strong spin-lattice coupling, the local dynamical fluctuations of the nuclear density in the critical-temperature region must exist and correlate with the local dynamical fluctuations of the magnetization.

In this letter, we report detailed information on the nuclear-magnetic interference observed in critical Small Angle Polarized Neutron Scattering (SAPNS) from $Fe_{65}Ni_{35}$. We interpret the observed interference as a result of the alignment of the spins inside the magnetic critical fluctuations leading to the local expansion of the lattice. The intensity of the scattering is determined by the nuclear and magnetic contrasts. Since the nuclear scattering length of the natural elements $b_{\mathrm{Fe}} = 0.98 \times 10^{-12}$ cm and $b_{\mathrm{Ni}} = 1.03 \times 10^{-12}$ cm are almost equal, then the nuclear contrast is provided mostly by the nuclear density fluctuations (number of atoms per unit volume). The magnetic contrast enhances, provided by a spin correlation in the critical fluctuations. Although an ordinary SANS at critical temperatures has a quasi-elastic nature and does not capture the dynamics of the magnetic fluctuation, the scattering allows one to unambiguously identify the shape of the scattering objects as one of the critical (long-living) fluctuations. The interference scattering reveals that the shape of the scattering nuclear objects is essentially the same as the magnetic one. This implies that the lifetime of the scattering nuclear object is correlated with, or terminated by, that of the magnetic fluctuation. Therefore, if the interference term in scattering appears, then the nuclear and magnetic fluctuations are correlated in the same (r,t)-space, which is the evidence for nuclear-magnetic coupling.

The nuclear-magnetic interference in SAPNS on invar was previously observed on an isotopic sample $Fe_{65}\,{}^{62}Ni_{35}$ at the temperature well below T_C [8, 9]. The isotope ^{62}Ni is a well-contrasting element on the background of the natural Fe atoms ($b_{\mathrm{Ni(62)}} = -0.87 \times 10^{-12}$ cm). The observed interference demonstrated the correlation of the inhomogeneous magnetic structure with the short-range fluctuations of the chemical composition. The interference based on the chemical inhomogeneities was not observed on the sample made of natural elements [9]. Another neutron interference experiment has recently been performed to observe the magneto-vibrational scattering showing coupling of spin system with phonon excitations [10].

The sample. – The classical invar $Fe_{65}Ni_{35}$ alloy, having FCC structure, was chosen for the study. In our previous papers [11–13] we had demonstrated the coexistence of two correlation length scales in a paramagnetic phase as the temperature approaches T_C. To interpret the appearance of the second correlation length, we introduce a position-dependent Curie temperature T_C, which fluctuates around an average value $\langle T_C \rangle$. The local T_C variations are described by the deviation of the transition temperature, ΔT_C, and by the characteristic length R_0. The experimentally determined parameters for $Fe_{65}Ni_{35}$ are: $\langle T_C \rangle = 485 \pm 0.5$ K, $\Delta T_C = 12.5 \pm 0.2$ K and $R_0 \approx 3 \times 10^3$ Å [13].

To demonstrate the invar properties of the sample we performed the measurements of the thermal expansion coefficient $\alpha = (1/l)(\mathrm{d}(\Delta l)/\mathrm{d}T)$ (see fig. 1). It nearly vanishes at the room temperature and increases smoothly as the temperature increases, having a strong change in the temperature range 470–530 K. In the paramagnetic region above 530 K it increases

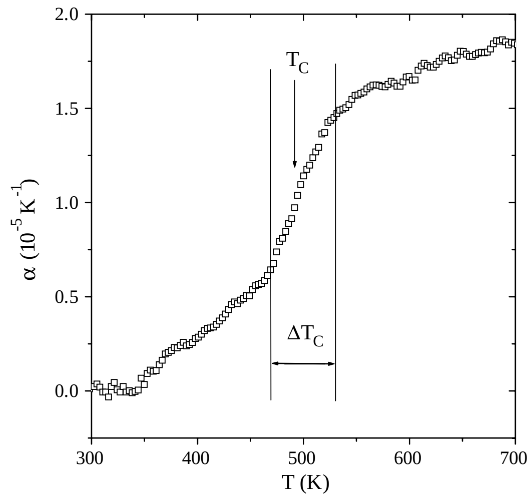

Fig. 1

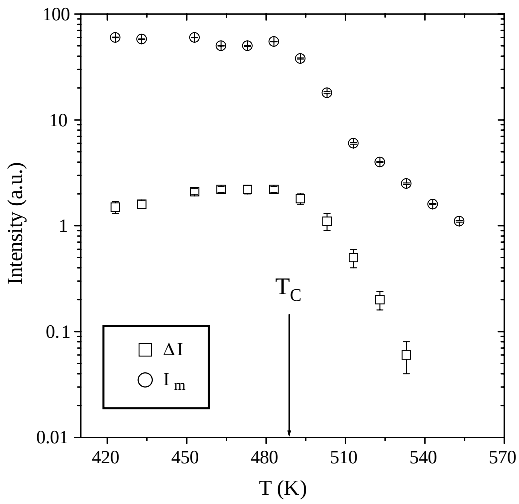

Fig. 2

Fig. 1 – Temperature dependence of the thermal expansion coefficient $\alpha = (1/l)(\mathrm{d}(\Delta l)/\mathrm{d}T)$.

Fig. 2 – Temperature dependence of the magnetic scattering intensity I_m and the symmetric polarization-dependent part of the scattering ΔI at $q = 0.01$ Å ($H = 70\,\mathrm{mT}$).

linearly. The strong change of α occurs at the critical range of the magnetic phase transition around $\langle T_\mathrm{C}\rangle = 485\,\mathrm{K}$.

Theory. – For SANS, in general, the cross-section of polarized neutrons consists of nuclear and magnetic contributions as well as the nuclear-magnetic interference contribution of the scattering:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = |A_\mathrm{n}|^2\mathcal{F}_\mathrm{n}(\vec{q}) + |A_\mathrm{m}|^2 m_\perp^2 \mathcal{F}_\mathrm{m}(\vec{q}) + 2\vec{P}_0\vec{m}_\perp\big(A_\mathrm{n}A_\mathrm{m}Re\mathcal{F}_\mathrm{nm}(\vec{q})\big), \tag{1}$$

where $A_\mathrm{n} = (bN_0)$ is a nuclear contrast with $b \simeq 10^{-12}\,\mathrm{cm}$ as an average nuclear scattering length, N_0 as an average atom density and $A_\mathrm{m} = rN_0$ is a magnetic contrast with $r = 0.54 \times 10^{-12}\,\mathrm{cm}$. $\vec{m}$ is the unit vector along the magnetization and $m_\perp = \vec{m} - (\vec{m}\vec{e})\vec{e}$ with $\vec{e} = \vec{q}/q$. The autocorrelation function of the nuclear density fluctuations can be given by $\mathcal{F}_\mathrm{n}(\vec{q}) = \int \mathrm{d}\vec{r} \exp[i\vec{q}\vec{r}]\langle W_\mathrm{n}(r)W_\mathrm{n}(0)\rangle$. The function $W_\mathrm{n}(r)$ describes the probability of the fluctuation of the nuclear density. Similarly, one can write for the autocorrelation function of the spin density fluctuations: $\mathcal{F}_\mathrm{m}(\vec{q}) = \int \mathrm{d}\vec{r} \exp[i\vec{q}\vec{r}]S_\mathrm{z}^2\langle W_\mathrm{m}(r)W_\mathrm{m}(0)\rangle$, where S_z is the z-component of the atom's spin. The function $W_\mathrm{m}(r)$ describes the probability of the spin collinearity along the magnetic filed (the z-axis). The cross-correlator of the nuclear-magnetic density fluctuations is given by $\mathcal{F}_\mathrm{nm}(\vec{q}) = \int \mathrm{d}\vec{r} \exp[i\vec{q}\vec{r}]\langle W_\mathrm{n}(r)W_\mathrm{m}(0)\rangle\langle S_\mathrm{z}\rangle$. In a particular case of the magnetic phase transition with a strong spin-lattice coupling, the amplitude of the magnetic critical fluctuations W_m is large. At the same time W_n is small. Therefore, the nuclear contribution to the scattering (proportional to W_n^2) is negligibly small compared to the magnetic one (proportional to W_m^2). Fortunately, the interference scattering is linear in W_n and then it is much larger than the pure nuclear contribution.

Experimental. – The SANS experiments were carried out at the SANS-2 scattering facility of the FRG-1 research reactor in Geesthacht (Germany). A beam of polarized neutrons with initial polarization $P_0 = 0.9$, wavelength $\lambda = 5.6$ Å ($\Delta\lambda/\lambda = 0.1$) and divergence of 1.5 mrad was used. The scattered neutrons were detected by a position-sensitive detector (128×128 pixels) within angular range ± 80 mrad except the very center of the detector which is shadowed by the beam-stop. The scattering intensity was measured in the temperature range from $T = 400$ K to $T = 600$ K with a step of 10 K. The temperature was well stabilized within 1 K. The external magnetic field from 7 to 70 mT was applied parallel to the sample's long dimension at an angle of 45° with respect to the incident beam (in the so-called "inclined" geometry). The sample was saturated magnetically by the applied field, so that the polarization of the transmitted beam had been changed with increasing the field from 0.95 to 1.0 of the initial polarization P_0. Following the standard procedure we determine the q-dependence of pure magnetic scattering by subtracting from the measured intensities the nonmagnetic background at $T \gg T_C$: $I_m(q,T) = I(q,T) - I(q,600)$. The polarization-dependent part of the scattering is determined as a difference $\Delta I(q) = I^+(q) - I^-(q)$ of the intensities for neutrons polarized parallel (+) and anti-parallel (−) to the magnetic field. The CoFe mirror was used as a polarizer. The sum of the nuclear and magnetic scattering amplitudes causes the reflection of the neutrons with the polarization parallel to the magnetic field. The magnetic field guides the neutron polarization to the sample. In analogy with the action of the polarizer, the interference scattering in the sample is additive (subtractive) for the reflected beam of polarized neutrons if the positive (negative) variations of the nuclear amplitude coincide with magnetic variations aligned by the applied magnetic field. Use of this "inclined" geometry allows us to observe the dynamic chiral (DC) contribution to the polarization-dependent part of the scattering [14–18] in addition to the nuclear-magnetic interference. The DC contribution is anti-symmetric with respect to the scattering angle, while the nuclear-magnetic interference term is purely symmetric. The anti-symmetric DC contribution vanishes after an averaging of $\Delta I(q)$ over the detector at $|q| = \text{const}$, and only the symmetric nuclear magnetic interference term survives. The data on the DC contribution near T_C will be published elsewhere.

Results. – Figure 2 shows the magnetic scattering intensity I_m and the absolute value of the "interference" intensity $|\Delta I(q)|$ at $q = 0.01$ and $H = 70$ mT as a function of temperature. I_m increases with decreasing temperature and saturates at $T = \langle T_C \rangle \approx 485$ K. ΔI becomes observable at $T \leq 530$ K and increases as T approaches $\langle T_C \rangle$. Then, it tends to decrease below 450 K and diminishes at $T < 400$ K. This temperature behavior of the interference term correlates with the strong change of the thermal expansion coefficient α at $T \sim T_C$ (fig. 1).

We observed a "negative" sign of the interference term, which corresponds to the opposite sings of the magnetic and nuclear contrasts. It is possible if the increasing magnetic contrast leads to decreasing the nuclear one, *i.e.* diminishing the local nuclear density. The experimental ratio $\Delta I/I_m$ is of order of 4–5% (fig. 2). Theoretically, we estimate it as $\Delta I/I_m \simeq (A_n A_m/A_m^2)(W_n W_m/W_m^2) \simeq (b/r)(W_n/W_m)$. Assuming that near T_C $|W_m| \sim 1$ and $W_n \simeq (-3\delta a/a)$, where a is a lattice constant, the ratio $\Delta I/I_m \simeq 10(-\delta a/a)$. Therefore, the corresponding expansion of the lattice is $(\delta a/a) \approx 0.004$. From the data presented below, we conclude that the range of this expansion is of order 50 Å and depends on T and H.

The q-dependence of the magnetic scattering intensity I_m is well described by the Lorentzian $I_m(q) = Z_m(q^2 + \kappa_c^2)^{-1}$, where $\kappa_c = R_c^{-1}$ is the inverse correlation length. This shape of the magnetic form factor corresponds to the decreasing correlation function in real space as $r^{-1}\exp[-\kappa_c r]$. The parameters Z_m and κ_c^2 have been obtained from the fit. According to [1], the mechanism of the spin-lattice coupling is such that the collinearity of the spins determines the expansion of a lattice or increase in volume occupied by atoms. From the shape of the magnetic correlation we know that the collinearity of the spins decreases as $1/r$ at $r \leq R_c$. This implies that $R_n \leq R_c$, where R_n is the correlation length of the nuclear-density fluctuation.

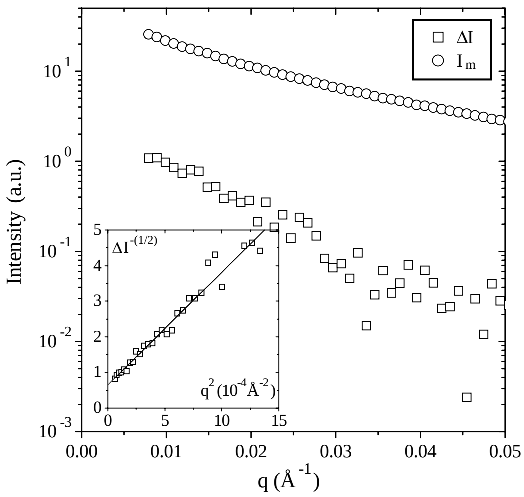

Fig. 3

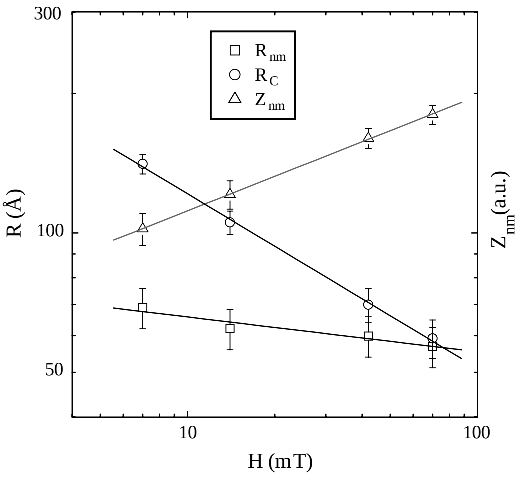

Fig. 4

Fig. 3 – q-dependence of the magnetic I_{m} and interference scattering ΔI at $T \simeq \langle T_{\mathrm{C}} \rangle$ and $H = 7\,\mathrm{mT}$. The inset demonstrates the linear dependence of $(\Delta I)^{-(1/2)}$ on q^2.

Fig. 4 – Magnetic-field dependence of the correlation lengths R_{c}, R_{nm} and the parameter Z_{nm} at $T \simeq \langle T_{\mathrm{C}} \rangle$.

Furthermore, we denote the volume inside the nuclear-magnetic fluctuations contributing to the interference scattering as an "interfering" one. This volume is limited by the interference correlation length R_{nm}, which determines, by supposition, the exponential decay of the nuclear-magnetic correlation $\exp[-r/R_{\mathrm{nm}}]$. The corresponding form factor of the interference term in q-space is a squared Lorentzian: $\mathcal{F}_{\mathrm{nm}}(q) = Z_{\mathrm{nm}}/(q^2 + \kappa_{\mathrm{nm}}^2)^2$, where $\kappa_{\mathrm{nm}} = R_{\mathrm{nm}}^{-1}$. We used this function for fitting the data of $\Delta I(q)$, and, as a result, two parameters Z_{nm} and κ_{mn} were obtained. The typical q-dependence of I_{m} and $\Delta I(q)$ for $T = 483\,\mathrm{K}$ and $H = 7\,\mathrm{mT}$ is displayed in fig. 3. The square root of the inverse intensity $(\Delta I)^{-1/2}$ is plotted in the inset of fig. 3 as a function of q^2. The obtained linear dependence verifies applicability of the squared Lorentzian as a form factor for the interference term. It also proves that the shape of the scattering nuclear objects is essentially the same as the magnetic one. This fact may be interpreted as if the nuclear fluctuation is correlated with, or caused by, the magnetic fluctuation.

The effect of the magnetic field on the correlation lengths R_{c} and R_{nm} at $T = 483\,\mathrm{K}$ is shown in fig. 4. The value of R_{c} decreases with the field. As is well known, the magnetic field renormalizes the behavior of the magnetic system in the vicinity of the transition temperature T_{C}. In this case the scaling is determined by not the reduced temperature $\tau = (T - T_{\mathrm{C}})/T_{\mathrm{C}}$ but by the magnetic field [19]. The observed behavior of R_{c} is a result of a crossover to the strong-field regime, which is determined by the condition $T(a_0/R_{\mathrm{c}}(H))^z = g\mu H$, where $z \simeq 5/2$ and a_0 is a constant of order of $1\,\text{Å}$. The fit gives value of the parameter $1/z = 0.38 \pm 0.01$, which is very close to the theoretical value equal of $2/5$, and $a_0 = 1.40 \pm 0.05\,\text{Å}$. The correlation length R_{nm} is smaller than R_{c} at small fields, and they become equal at the large applied field of $H = 70\,\mathrm{mT}$. Such a dependence is similar for different temperatures around T_{C}. Therefore, we can conclude that the interfering volume is limited by R_{n} at the small field and by R_{c} at the large one. We suppose that the magnetic field may affect the interference form factor $\mathcal{F}_{\mathrm{mn}}$ in two opposite ways. First, it suppresses the critical fluctuations of the magnetization,

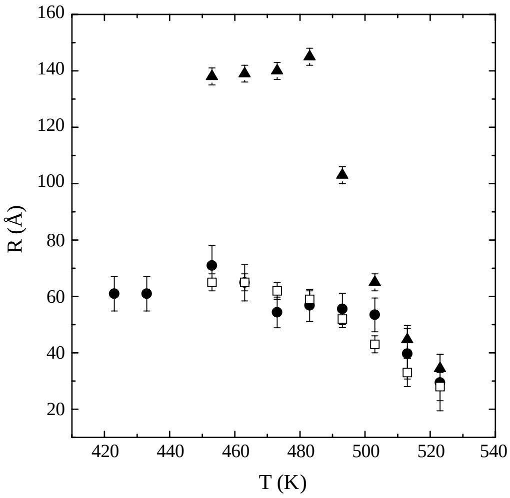

Fig. 5 – Temperature dependence of the correlation lengths R_c on the magnetic field $H = 7$ (triangles) and 70 mT (open squares) and R_{nm} in $H = 70$ mT (closed circles).

which results in decreasing the interfering volume and, therefore, R_{nm}. On the other hand, the magnetic field increases the alignment of the spin inside the fluctuations resulting in an increase of the average spin $\langle S_z \rangle$ and, therefore, the intensity of the interference term Z_{nm}. According to the scaling theory (see, for example, [19]), $Z_{nm} \sim \langle S_z(H) \rangle \sim (\kappa_c(H))^{1/2} \sim H^{1/5}$. The magnetic-field dependence of Z_{nm} for $T = 483$ K is shown in fig. 4. Z_{nm} grows with magnetic field as H^α, where $\alpha = 0.25 \pm 0.05$, which is close to the theoretical value. The amplitude Z_m does not depend on H.

Figure 5 shows the temperature dependence of the correlation lengths R_c at two different magnetic fields: $H = 7$ mT (triangles) and 70 mT (open squares) and R_{nm} at $H = 70$ mT (closed circles). When the field is small, the correlation length R_c increases with lowering temperature and is saturated at $T \approx \langle T_C \rangle = 485$ K. If the field is large ($R_c > R_c(H)$), the strong-field regime takes place and the T-dependence of critical fluctuations tends to disappear. The temperature dependence of R_{nm} at a magnetic field of 70 mT coincides with R_c in the whole temperature range.

Conclusion. – The nuclear-magnetic interference in SAPNS from invar $Fe_{65}Ni_{35}$ is caused by the spin-lattice coupling in the critical region for the ferromagnetic transition. It reveals the correlation between critical fluctuations of magnetization and fluctuations of the nuclear density. The observation of spin-lattice coupling inside the critical fluctuation may provide a basis for further theoretical models.

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The authors are grateful to Drs V. V. RUNOV, G. P. GORDEEV and N. H. VAN DIJK for stimulating discussions and to N. GEERLOFS for the thermal expansion measurements. The authors would like to thank RFFR (projects SS-1671.2003.2 and 01-02-17286), Russian program "Quantum microphysics" and the Russian State Program "Neutron Research of Solids". PNPI-team acknowledges GKSS-Geesthacht for hospitality.

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