

Influence of dipolar interactions on the critical dynamics in nickel

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The study of critical phenomena has always been deeply connected with magnetism in solid state materials. Nickel, one of the three archetypical room temperature ferromagnets, is understood to be an itinerant system with a high degree of localization regarding its magnetically active electron states. We report a high-resolution neutron spectroscopy investigation of nickel (Ni) near its Curie temperature T_C , extending measurements to unprecedentedly small wave vectors $q_{\min} = 6.6 \times 10^{-3} \text{ \AA}^{-1}$ and achieving an energy resolution of $\Delta E = 4.9 \text{ \mu eV}$. Our analysis of the spin-wave dispersion reveals that the dipolar wave vector $q_D = 6.4 \times 10^{-3} \text{ \AA}^{-1}$ is approximately half of the previously reported value, but strictly nonzero to rationalize the observed excitations. The spin-wave stiffness $D(T)$ is in good agreement with the literature, but most importantly, we uncover evidence that the linewidth of the spin waves follows the dynamical scaling characteristic of a dipolar ferromagnet rather than an isotropic one. In contrast, the linewidth of the fluctuations above T_C exhibit scaling consistent with the Résibois-Piette function and renormalization-group predictions, despite the itinerant ferromagnetic nature of Ni. This observation suggests a pronounced localization of the $3d$ electrons responsible for magnetic scattering.

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I. INTRODUCTION

Ferromagnetic Ni has been thoroughly investigated using neutron scattering techniques since the late 1960s [1–7]. Being a metal, Ni is an excellent representative of an *itinerant* ferromagnet, in which, as opposed to localized electron systems, the electronic band structure determines the magnetic behavior. On this scale, materials can be classified by comparing their magnetic stiffness constant D extracted from the low- q spin-wave dispersion to their Curie temperature T_C . Note that D is essentially proportional to the exchange interaction J . Localized systems, as understood in terms of the atomistic Heisenberg model, achieve $D/k_B T_C \approx 1 \text{ \AA}^2$, while a high degree of itinerancy leads to $D/k_B T_C \gg 1 \text{ \AA}^2$. Here we have used the D values at $0.8 T_C$ from Shirane [8]. Ni scores high on the itinerancy scale as summarized by, e.g., Endoh and Böni [9].

This classification quantifies the deviation of a particular magnet from the classical Heisenberg model, meaning that influences attributed to the band structure are significant for large ratios $D/k_B T_C$. Generally, the dynamics of localized and itinerant ferromagnets such as EuO [10–13], EuS [10,11,14], and Pd₂MnSn [15,16] on the one hand, and Fe [17–19], Ni

[20,21], and MnSi [22] on the other hand, are well characterized thanks to the many neutron scattering studies.

Interestingly, in the low- q and small- E region in the ferromagnetic as well as the paramagnetic phase and close to T_C , all of these systems are well understood in terms of a Hamiltonian based on the Heisenberg model. This understanding also extends to the incorporation of weak, long-range dipolar interactions for localized systems. In itinerant systems, more complex behavior is observed in the vicinity of T_C due to additional scattering channels for the spin fluctuations. Exemplarily in Fe, the critical dynamics has been found to differ from Heisenberg-like behavior since the Fermi surface smears out significantly at high temperatures, thus giving rise to low- q spin-flip excitations of the conduction electrons [18].

Ferromagnetic Ni lies at the upper end of the itinerancy scale, making it a suitable candidate to more clearly identify effects due to the influence of the conduction electrons on the spin dynamics. Investigations of Ni have proven challenging due to the large spin-wave stiffness $D \approx 400 \text{ meV \AA}^2$ at room temperature. As the spin-wave stiffness renormalizes close to the Curie temperature, Ni offers the unique possibility to probe long-wavelength spin waves below T_C and paramagnetic fluctuations above T_C to study the influence of long-range interactions such as the dipolar coupling and itinerant correlations over the length scale of a large number of unit cells. As the dipolar wave number $q_D \propto D^{-1/2}$, q_D becomes particularly small in nickel and signatures of dipolar interactions will appear only at $q \leq q_D$, thus requiring exceptionally good momentum and energy resolution.

The direct observation of dipolar coupling in the spin-wave dispersion or in the paramagnetic fluctuations has been elusive. The neutron resonance spin-echo spectrometer RESEDA

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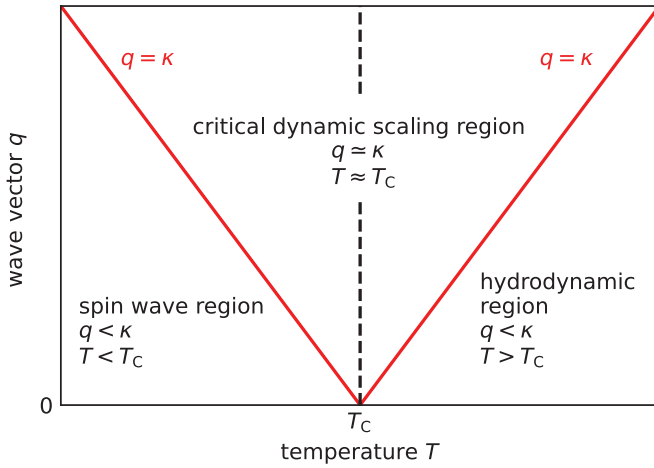


FIG. 1. Sketch of the wave vector transfer vs temperature diagram near the Curie temperature T_C . The red lines given by $q = \kappa$ define three regions of different asymptotic behavior. Corresponding to their names, a magnetic system exhibits collective excitations in the spin-wave region, whereas spin diffusion is dominant in the hydrodynamic region. According to dynamical scaling theory, the asymptotic forms of the regions merge at the boundaries $q = \kappa$.

at FRM II, when set up in the MIEZE configuration is uniquely suited for the investigation of the critical magnetic scattering thanks to its high momentum and energy resolution. Therefore, we have studied Ni in the temperature range $T_C - 2 \text{ K} \leq T \leq T_C + 8 \text{ K}$, where $T_C = 627 \text{ K}$. We find that our results are explained best by incorporating dipolar interactions.

We review the dynamical scaling theory in the context of ferromagnetism in Sec. II. The basics of the neutron spin-echo method, the experimental setup, the data reduction, and the data processing are summarized in Sec. III. The data analysis in the context of a dipolar spin-wave dispersion and dynamical scaling is presented in Sec. IV, while Sec. V discusses the results in the light of previous studies of ferromagnets.

II. THEORY OF CRITICAL DYNAMICS OF HEISENBERG FERROMAGNETS

Fundamental to the theory of critical phenomena and scaling is the existence of a correlation function $C_\xi^{\mathbf{M}}(\mathbf{r}, t)$ of the magnetization operator $\hat{\mathbf{M}}$ and its associated correlation length ξ , which approaches infinity at the transition temperature T_C . Simultaneously, ξ can be considered to be a measure of the temperature deviation from T_C such that $\xi = \xi_0 t^{\nu(\nu')}$ with the reduced temperature $t = |T - T_C|/T_C$ and $\nu, (\nu')$ the critical exponents above (below) T_C [23]. With regard to magnetism, $C_\xi^{\mathbf{M}}$ is related to the spin-spin correlation function $\langle S_i^\alpha S_j^\beta \rangle$, which fully encapsulates the static magnetic structure and its dynamics.

In momentum and temperature space, the inverse correlation length $\kappa = \xi^{-1}$ defines three separate regions for $T \cong T_C$. Figure 1 shows the separation into the *spin-wave region* ($q < \kappa, T < T_C$), the *critical dynamic scaling region* ($q \simeq \kappa, T \approx T_C$), and the *hydrodynamic region* ($q < \kappa, T > T_C$). Within each region, the investigated system exhibits a different dynamical

behavior. The dynamical behavior of a system deep in the ferromagnetic state ($T \ll T_C$) and far in the paramagnetic phase ($T \gg T_C$) and consequently its observed response to neutron scattering (NS) is known from spin-wave theory and statistical mechanics, respectively. Dynamical scaling theory (DST) extends this description and allows one to predict the renormalization of the spin-wave dispersion and the lifetime of paramagnetic fluctuations close to T_C .

A. Spin-wave region

The spin waves in nickel can be described by a microscopic model of exchange coupled spins with dipolar anisotropy in an external magnetic field $\mathbf{H}$ along the z axis. The corresponding Hamiltonian reads $\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{dip}} + \mathcal{H}_{\text{Zeeman}}$. Holstein and Primakoff [24] calculated the spin-wave dispersion on the basis of the model for an isotropic ferromagnet to be

$$E_{\text{SW}}(\mathbf{q}) = [(Dq^2 + g\mu_0\mu_B H)(Dq^2 + g\mu_0\mu_B H + g\mu_0\mu_B M(T, H) \sin^2(\theta_{\mathbf{q}}))]^{1/2} \quad (1)$$

for small $\mathbf{q}$ values. Here, D is the material specific *spin-wave stiffness*, μ_B the Bohr magneton, $M(T, H)$ the magnetization, g the Landé factor ($g = 2.2$ for Ni [25]), and $\theta_{\mathbf{q}}$ the angle between the magnetization direction and the momentum transfer $\mathbf{q}$. Specifically for $H = 0$, the Zeeman term vanishes, and in the demagnetized state, the magnetic domains can be assumed to be randomly oriented. Then the average of $\theta_{\mathbf{q}}$ over all domains yields $\langle \sin^2(\theta_{\mathbf{q}}) \rangle = \frac{2}{3}$. Therefore, the dispersion becomes isotropic, and the measured spin-wave energy only depends on the magnitude of the wave vector transfer $\mathbf{q}$.

The treatment of spin waves in a ferromagnetic material assumes a fully ordered magnetic state, such that spin-wave stiffness D_0 and magnetization M_{sat} essentially represent the quantities at $T = 0$. The commonly known temperature-dependent decrease of the saturation magnetization $M(T)/M(0) \propto 1 - aT^{3/2}$, referred to as Bloch's law, is a consequence of spin-wave excitations above absolute zero. This law breaks down close to T_C . Here, critical scaling theory addresses the temperature dependence of these quantities as follows:

$$M(T) = M' |t|^\beta, \quad (2)$$

$$D(T) = D' |t|^\mu, \quad (3)$$

where β designates the critical exponent for the magnetization and $\mu = \nu' - \beta$ the exponent for the spin-wave stiffness [23,26] as derived from critical scaling theory. The introduction of the *primed* quantities M' and D' highlights that they do not correspond to the $T = 0$ values because the scaling behavior is only valid close to T_C . By rewriting the dipolar contribution to the dispersion as

$$\frac{2}{3} g\mu_0\mu_B M(T) = D(T) q_D^2, \quad (4)$$

the dipolar wave number q_D emerges as a material-dependent parametrization of the dipolar coupling strength. Moreover, q_D is essentially temperature independent as $\mu \simeq \beta$.

Approaching T_C from below, the spin-wave stiffness and the magnetization vanish. The self-interaction of the plethora of low-energy spin-wave states leads to their decay resulting in a finite lifetime $\hbar/\Gamma$. The broadening of the spin-wave

excitations can be directly measured by inelastic neutron scattering. Furthermore, the line shape of the excitations must be considered in order to properly disentangle the intrinsic shape from the instrumental broadening of the signal. The line shapes of the spin waves are modeled in a first approximation as Lorentzian distributions in energy, where the half width at half maximum is given by $\Gamma(q)$.

B. Critical dynamic scaling region

As assumed in the framework of dynamical scaling, a system is fully characterized by its behavior in the three regions, depicted in Fig. 1. Any dominating mode, such as a spin wave, will continue arbitrarily close to the critical temperature T_C without changing its fundamental character. At the critical point, i.e., T_C , ξ diverges, and the so-far valid description breaks down.

Besides this singularity, the dynamical scaling hypothesis rests on the assumption that anywhere else in the (q, T) plane ($q \neq 0$), the correlation function varies smoothly. At the borders $q = \kappa$ separating the regions, the mathematical description of the system asymptotically matches. Here, matching refers to a generalized concept of a characteristic frequency $\omega_c(q)$, which bridges the distinction between the frequency of a spin-wave excitation in the ordered state and the linewidth of a quasielastic spin-diffusion process in the disordered state. The characteristic frequency is defined by

$$\int_{-\omega_c}^{+\omega_c} f_{q\xi}(\omega) d\omega = \frac{1}{2}, \quad (5)$$

where $f_{q\xi}(\omega)$ designates the line shape function. According to scaling theory, the characteristic frequency of the magnetization of an isotropic ferromagnet in three dimensions [26] is expected to follow

$$\omega_c(q) = Dq^2 = \tilde{D} \sqrt{\frac{\kappa}{q}} q^{5/2} \simeq \Lambda q^{(5-\eta)/2}, \quad (6)$$

where Λ determines the frequency scale of the decaying fluctuations and $D = \tilde{D}\sqrt{\kappa}$ designates the stiffness constant. The Fisher exponent $\eta = 0.034$ is usually neglected because it is so small. With the mathematical expression of the dynamical scaling hypothesis in terms of the $f_{q\xi}$ and $\omega_c(q)$ and their scaling properties with respect to the correlation length ξ , it becomes experimentally testable, for example, in scattering experiments [26].

Renormalization-group theory allows the calculation of the shape function at finite q in the limit of $T \rightarrow T_C$ ($\xi \rightarrow \infty$). The analytical representation is given by

$$f_{q\xi=\infty}(q, s) = \frac{1}{\pi \Lambda q^{5/2}} \operatorname{Re} \left\{ \frac{1}{is + \alpha[1 + i(\alpha/a)s]^{-0.6}} \right\}, \quad (7)$$

where $\operatorname{Re}\{\dots\}$ returns the real part of a complex function, where $s = \omega/\omega_c$, $\alpha = 0.78$, and $a = 0.46$ [21,27]. Deep in the hydrodynamic region, the function becomes a Lorentzian.

C. Hydrodynamic region

The hydrodynamic region (Fig. 1) of a ferromagnet largely coincides with its paramagnetic state, where the dynamical scaling hypothesis persists, predicting the inverse lifetime to be $\Gamma(q) = \Lambda\gamma(\kappa/q)q^{5/2}$. Here, $\gamma(\kappa/q)$ is referred to as the *dynamical scaling function*, which contains the entire

temperature dependence in terms of the inverse correlation length κ [26]. While dynamical scaling theory constrains the properties of $\gamma(\kappa/q)$ by requiring it to be a homogeneous function, which allows the determination of its asymptotic behavior for $\kappa \rightarrow 0$, the form at any other value of the scaled variable $x = \kappa/q$ is not known from the outset.

Using a mode-coupling (MC) approach for the treatment of the time-dependent spin correlation function, Résibois and Piette deduced an integral equation for $\gamma(x)$ of an isotropic Heisenberg system that satisfies the asymptotic scaling behavior [28]. Similarly, Iro found a solution for $\gamma(x)$ via renormalization-group theory calculations, which in addition yielded an expression for the dynamical shape function $f_{q\xi}(q, \omega)$ [29].

Experimental results gathered on typical ferromagnets such as Fe [17], Co [30], Ni [20,21], EuS [31], and EuO [12,32] supported the general notion of the dynamical scaling hypotheses and the prediction of the dynamical scaling function by Résibois and Piette. Discrepancies, however, showed up in systems EuO [10] and EuS [10,31], which are known to exhibit significant dipolar coupling besides the dominant exchange interaction [14]. To understand these discrepancies, Frey and Schwabl developed a theoretical framework for calculating the dynamical scaling function in the presence of dipolar interactions based on a mode-coupling ansatz [33–35].

As a result, the dipolar wave number enters the dynamical scaling function in terms of an additional scaling variable q_D/κ . The anisotropy of the dipolar interactions, furthermore, lifts the degeneracy of the *longitudinal* (L) and *transversal* (T) fluctuations leading to $\Gamma^\alpha(q) = \Lambda\gamma^\alpha(\kappa/q, q_D/\kappa)q^{5/2}$ with ($\alpha = L, T$) [33–35].

III. EXPERIMENT AND DATA REDUCTION

A. MIEZE technique

Since their inception by Mezei [36], and Gähler *et al.* [37], Larmor precession techniques have been at the forefront of the investigation of slow magnetic dynamics, with unprecedented E resolution. The modulation of intensity with zero effort (MIEZE) technique especially lends itself to the study of magnetic fluctuations in ferromagnetic materials since it is not affected by magnetic samples and magnetic field environments. While conventional spin-echo techniques are generally susceptible to depolarizing conditions, MIEZE performs the spin manipulations upstream of the sample position, which is why depolarization at the sample position does not prevent the measurement.

As depicted in Fig. 2, to employ the MIEZE technique the neutron beam gets polarized by the polarizer P_1 . Two radio-frequency spin flippers (RF flippers) are tuned to different frequencies $f_{A,B}$ and are placed downstream of P_1 with a distance L_{AB} between them. After passing the spin analyzer P_2 the manipulation of the neutron spin by the RF flippers RF_A and RF_B has created a neutron beam with a sinusoidal intensity modulation $I = y_0 + A \cos(2\pi f_M + \phi)$ at the detector position. This oscillation has a frequency of $f_M = 2(f_B - f_A)$ and can be recorded with a time-resolving detector D behind the sample S if the *MIEZE condition* is fulfilled. The *MIEZE condition*,

$$f_B/f_A = (L_{AB} + L_{BD})/L_{BD}, \quad (8)$$

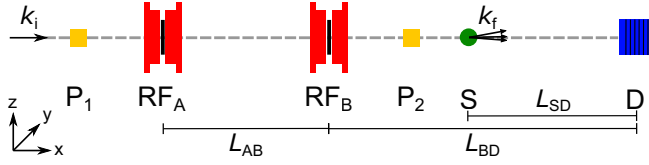


FIG. 2. Sketch of the MIEZE setup implemented at the RESEDA beamline. The spin state of the incoming neutrons (k_i) is selected at the polarizer (P_1). When the neutron beam passes the radio-frequency spin flippers ($RF_{A,B}$) and the spin analyzer (P_2) a sinusoidal intensity modulation is created at the echo point. This modulation with frequency $f_M = 2(f_B - f_A)$ can be measured with a time-resolving CASCADE detector (D). If neutrons are inelastically scattered by the sample (S), the ratio of amplitude to mean value decreases compared to the unperturbed, modulated beam.

relates the frequencies to the distances between the RF flippers and the detector position. L_{BD} is the distance between RF_B and the detector. Each frequency corresponds to a Fourier time (MIEZE or spin-echo time)

$$\tau_M = \frac{h}{m_n v_i^3} f_M L_{SD}, \quad (9)$$

where v_i is the velocity of the incoming neutrons and L_{SD} is the distance between the sample and the detector. A MIEZE measurement provides information about the dynamics of the sample in terms of the contrast $C = A/y_0$, which is the ratio of the amplitude A of the oscillation and the mean value y_0 of the intensity pattern. For a more detailed description of the MIEZE technique see Ref. [38].

The contrast is the equivalent of the polarization measured in conventional neutron spin-echo spectroscopy and is therefore related to the normalized intermediate scattering function $\text{Re}\{I(q, \tau_M)\}/S(q)$ [39,40]. Here, $I(q, \tau_M)$ and $S(q)$ designate the intermediate scattering function and the static structure factor, respectively, as provided by the Van Hove theory [41]. $\text{Re}\{I(q, \tau_M)\}$ can be computed as the cosine-Fourier transform of the dynamic structure factor $S(q, E)$ with respect to the energy transfer E , while $S(q)$ is the energy integrated $S(q, E)$. Using a MIEZE instrument with a two-dimensional (2D) detector and energy-dependent detection efficiency,¹ it has been successfully demonstrated that a comprehensive analysis of MIEZE data requires us to go beyond the idealized equivalency of the contrast and the normalized intermediate scattering function [42–44]. Therefore, in the analysis process we have calculated the MIEZE contrast by numerical methods utilizing the more involved expression

$$C(q, \tau_M) = \frac{\int_{>-E_i}^{\infty} \frac{k_f}{k_i} S_{\text{eff}}(q, E) \cos\left[2\pi f_M L_{SD} \left(\frac{1}{v_i} - \frac{1}{\sqrt{v_i^2 + 2E/m}}\right)\right] dE}{\int_{>-E_i}^{\infty} \frac{k_f}{k_i} S_{\text{eff}}(q, E) dE}, \quad (10)$$

¹The energy-dependent detection efficiency has been shared by the manufacturer of the CASCADE detector and is included in the MODEL MIEZELB package used for the data reduction.

which was introduced for proper consideration of scattering events with large energy transfer [44].

The numerator of Eq. (10) is analogous to $\text{Re}\{I(q, \tau_M)\}$ in the normalized intermediate scattering function, but introduces three changes to the simple cosine-Fourier transform applied to calculate $\text{Re}\{I(q, \tau_M)\}$. First, the argument in the cos term becomes nonlinear in E . Second, the lower integration boundary $-E_i$ takes into account that energy transfers $E > E_i$ from the neutron to the sample are not possible. For more details regarding the reasoning and consequences see Beddrich *et al.* [44]. Third, $S_{\text{eff}}(q, E) = \varepsilon(E)S(q, E)$ is introduced as the "effective" dynamical structure factor observed by the CASCADE detector and includes the energy-dependent detection efficiency $\varepsilon(E)$. Similarly, the denominator includes the same adjustments to normalize the contrast at $\tau_M = 0$ to 1.

For a sharply peaked dynamical structure factor and the relevant energy transfers being small with respect to the energy of the incoming neutrons, $E \ll E_i$, these extensions become negligible or factor out completely. Then the well known expression of the normalized intermediate scattering function is recovered. Aiming for the most comprehensive analysis we employed Eq. (10), since we need to be sensitive to subtle changes in the shape of $S(q, E)$ as explained in Sec. III C.

B. Experimental setup and data acquisition

The experiment was performed using the MIEZE spectrometer RESEDA at FRM II, which was set up in its standard small-angle neutron scattering configuration. Pre-tuned MIEZE times τ_M ranging from 10^{-4} to 1 ns were used. The triangular wavelength distribution of the velocity selector was centered at $\lambda_0 = 6.0 \text{ \AA}$ with a bandwidth of $\frac{\Delta\lambda}{\lambda} = 0.12$. Two square shape apertures of $10 \times 10 \text{ mm}^2$ over a 4.4 m distance collimated the beam in front of the sample and neutrons arriving at the sample from double reflections in the neutron guide have been removed using a 40 arcmin collimator.

The fast, time-resolving, position-sensitive detector (PSD), also referred to as CASCADE [45], was placed at a distance of $L_{SD} = 3.43 \text{ m}$ behind the sample position with an active detection area of $200 \times 200 \text{ mm}^2$. A circular beam stop before the detector blocked the direct beam (DB).

The sample used in the presented study was a highly enriched ^{64}Ni single crystal that is shaped like a droplet. Thus the nuclear incoherent scattering is small. The lattice constant of fcc Ni at room temperature and atmospheric pressure is $a = 3.52 \text{ \AA}$. The sample is 20 mm long and has a diameter of about 13 mm for the thicker part of the drop-shaped sample, allowing for a $10 \times 10 \text{ mm}^2$ beam spot. A thermocouple (type C) [46] in direct contact with the surface of the crystal served as a temperature sensor.

The experimental determination of T_C was performed by observing the critical magnetic scattering integrated in a region of interest (ROI) around the direct beam spot on the CASCADE detector. As a function of temperature, the intensity exhibits a maximum, as the magnetic susceptibility $\chi(T)$ diverges at T_C . The temperature was ramped with a rate of 0.2 K/min and a neutron counting time of 30 s per point was used. Several T sweeps across T_C showed no hysteresis and confirmed temperature stability.

MIEZE data were acquired after adjusting the sample temperature to a value $T = T_C + \Delta T$ with $\Delta T \in [-2 \text{ K}, -1 \text{ K}, +0 \text{ K}, +1 \text{ K}, +2 \text{ K}, +4 \text{ K}, +8 \text{ K}]$. For each T and τ_M the counting time was set to 1 h, which achieves high enough counting statistics for a reliable contrast determination at large q values. Additionally, background data were recorded at $T = T_C + 30 \text{ K}$, where no magnetic scattering is observed anymore. Lastly, the time resolution of the instrument was measured with a graphite reference sample [38,47].

C. Data reduction

At RESEDA, data acquisition produces a five-dimensional dataset $I_{\text{dat}}(\tau_M, n, t_{\text{bin}}, x, y)$ [47]. The recorded parameters are the MIEZE time τ_M ; the index n that identifies one of the six boron foils, where a neutron was detected; and the index of the internal time binning of the CASCADE detector t_{bin} . Furthermore, each foil provides the position x and y of the detected neutron.

Similar to the investigation conducted by Säubert *et al.* [19], four processing steps were performed to transform the raw data into a tractable set of parameters consisting of momentum transfer q and energy transfer E :

- (i) The ROIs on the detector image are selected in which data processing occurs. The ROIs reflect the symmetry of the scattering signal.
- (ii) In each ROI the contrast as a function of τ_M is determined from the time-modulated intensity of the neutron signal.
- (iii) The total momentum transfer q is calculated from the pixels within the ROI: $C(\text{ROI}, \tau_M) \rightarrow C(q, \tau_M)$.
- (iv) The relevant physical quantities are extracted by fitting appropriate models to the $C(q, \tau_M)$ data.

Improvements have been made regarding the standardization of the data reduction procedure and to account for the low magnetic scattering intensity of Ni. In contrast to a previous study on Fe [19], the last point is addressed by selecting ROIs, which are parametrized as circular arcs. The radial coverage is increased with increasing scattering angle 2θ about the direct beam at the expense of q resolution. This way, the larger area of the ROI includes enough magnetic scattering intensity for a reliable contrast determination.

The software package MIEZEPY [47] is a dedicated tool for processing the MIEZE raw data and has been used to perform the data reduction steps (i) and (ii). The momentum transfer q is calculated under the assumption of elastic scattering, i.e., $q = \frac{4\pi}{\lambda} \sin \theta$. A more intricate description of all four reduction steps can be found in [43].

Equation (10) is applied to extract the relevant physical parameter, e.g., the linewidth Γ and the spin-wave energy E_{SW} , if applicable. The analysis distinguishes between three distinct functions $S(q, E)$, depending on the sample temperature, and is motivated by critical scaling theory (see Appendix A for more details). In the spin-wave region, a sum of two Lorentzian distributions located at $\pm E_{\text{SW}}(q)$ with a linewidth $\Gamma_{\text{SW}}(q)$ was used [Eq. (A1)]. Equation (7) was used for $T = T_C$. Above T_C , the line shape function, which Iro has derived based on asymptotic renormalization-group theory [Eq. (A2)], was employed for the extraction of the linewidth [48]. This approach has also been applied successfully by

Böni *et al.*, with the material parameters of the inverse correlation length $\kappa(t) = 0.81t^{0.701} \text{ \AA}^{-1}$ and the frequency scale $\Lambda = 350 \text{ meV \AA}^{2.5}$ at T_C [21]. After transforming these model functions into the time domain via Eq. (10), the free parameters are optimized using *least squares* minimization. During the data analysis, we checked the consistency of our results with the reported values, but refrained from cross validation with static scattering data that were initially used for the determination of T_C as the counting statistics was poor.

The result of this analysis is exemplarily shown in Fig. 3 for the different temperature regimes. The data on the left-hand side depict the contrast with increasing q (from blue to yellow). The data on the right-hand side show the corresponding $S(q, E)$ functions.

IV. ANALYSIS REGARDING SIGNATURES OF DIPOLAR INTERACTIONS

A. Spin-wave dispersion

Knowing the excitation energy of the spin waves, E_q , the spin-wave dispersion at $\Delta T = -1 \text{ K}$ and $\Delta T = -2 \text{ K}$ can be determined. The T dependence of the dispersion is shown in Fig. 4. The vertical and horizontal error bars correspond to the standard deviation σ_{E_q} of the excitation energy and the standard deviation σ_q of the q distribution given by the ROI on the PSD, respectively.

In order to compare the measured data points of the spin-wave dispersion with Holstein-Primakoff theory, the data have to be deconvoluted with respect to the q resolution of RESEDA. Hammouda and Mildner suggested representing the resolution as a normal distribution [49] and derived an analytical expression for its standard deviation. The main contributions arise from the broad, triangular wavelength spectrum and the divergence of the incident beam that is determined by the neutron guide and apertures in front of the sample. The measured dispersion is fitted using a convolution of Eq. (1) and the q -resolution function.

In total, six different models (M1–M6) have been compared with the data. They vary in the approach of estimating and constraining some of the free parameters within the spin-wave model. A detailed description of the constraints of the different models, as well as a comparison of the resulting fitting parameters, is given in Appendix B.

Here, we focus on the models M4 and M6 as they involve only one fitting parameter, namely, D' , that parametrizes the spin-wave stiffnesses in Eq. (3), i.e., the strength of the exchange interactions. The associated critical exponent $\mu = 0.3407$ is fixed using the field theoretical result for three-dimensional (3D) ferromagnets [50]. M6 represents the model neglecting any dipolar interaction, while M4 incorporates dipolar interactions from experimental magnetization data published by Neugebauer [51]. The magnetization is directly put into Eq. (1). The extracted spin-wave stiffness parameters are $D' = 431.1 \pm 10.0 \text{ meV \AA}^2$ (Heisenberg, dipolar) and $D' = 475.4 \pm 16.1 \text{ meV \AA}^2$ (pure Heisenberg). The result of the fits is shown in Fig. 4 by the red solid and the blue broken lines, respectively.

The blue broken lines represent the model neglecting the dipolar interactions, i.e., $q_D = 0$. In contrast,

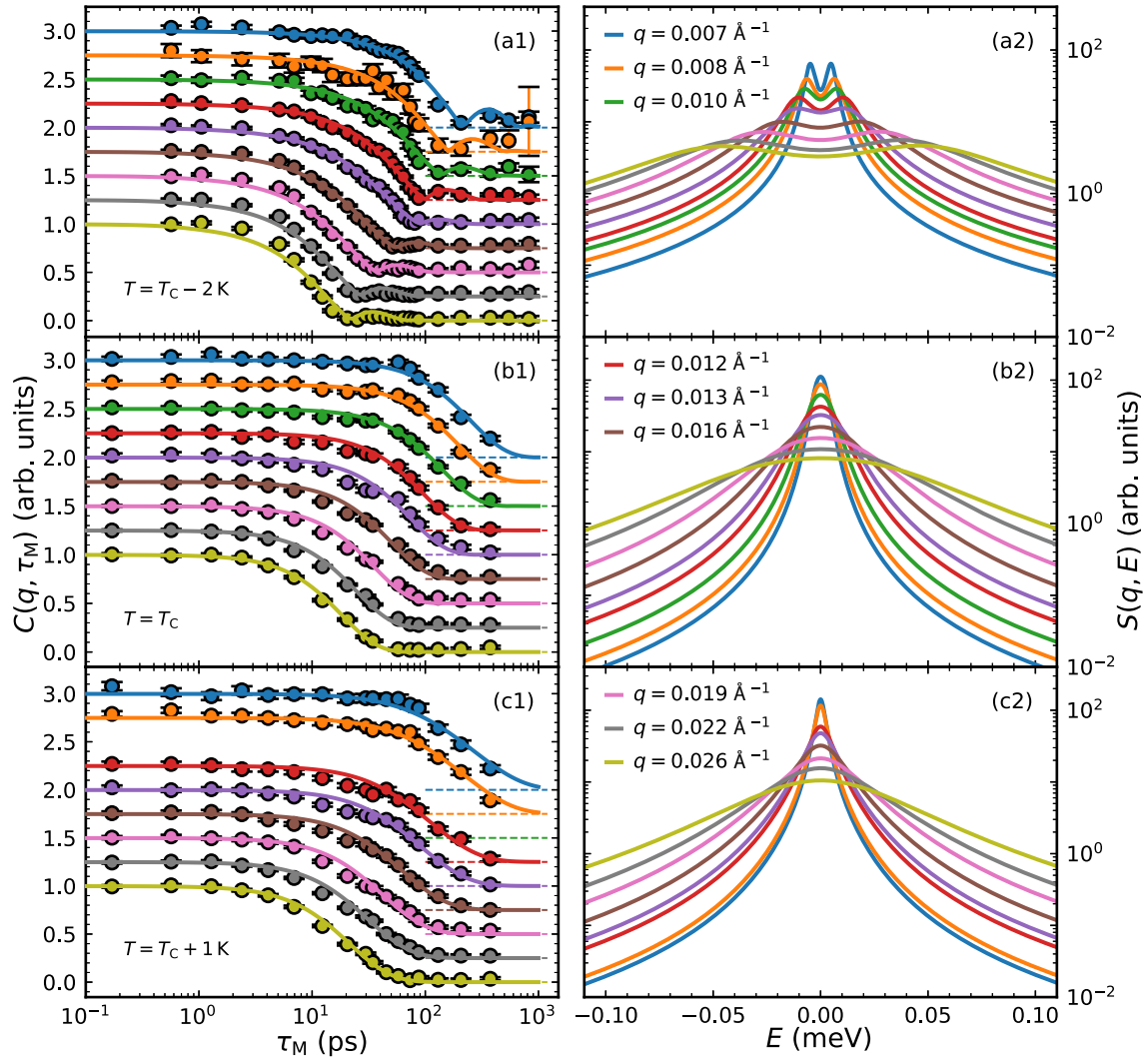


FIG. 3. The panels (a1), (b1), and (c1) on the left-hand side show the contrast for the temperatures $T_C - 2$ K, T_C , and $T_C + 1$ K, respectively. The curves are shifted vertically by an offset for a clear presentation and their corresponding 0 is indicated by a broken line. The top curve (blue) corresponds to the smallest wave vector $q = 0.007 \text{ \AA}^{-1}$. The solid lines show the best fit to the data and were calculated using Eq. (10) using the appropriate shape function. The corresponding line shape functions are shown on the right-hand side in panels (a2), (b2), and (c2).

the red solid lines visualize a fit with $E_{\text{dip}}(\Delta T = -2 \text{ K}) = 3.2 \mu\text{eV}$ and $E_{\text{dip}}(\Delta T = -1 \text{ K}) = 1.6 \mu\text{eV}$ corresponding to $q_D(\Delta T = -2 \text{ K}) = 7.2 \times 10^{-3} \text{ \AA}^{-1}$ and $q_D(\Delta T = -1 \text{ K}) = 5.7 \times 10^{-3} \text{ \AA}^{-1}$, respectively. The comparison of the χ_{red}^2 values of the two models clearly shows that the dipolar model ($\chi_{\text{red}}^2 = 3.1$) provides a more appropriate description than a pure Heisenberg model ($\chi_{\text{red}}^2 = 7.9$).

The main difference between the models lies in their agreement or disagreement with the data points $q \leq 0.01 \text{ \AA}^{-1}$, which is a result of the dipolar contribution of the order of $q_D \approx 0.006 \text{ \AA}^{-1}$. It indicates the crossover from a quadratic to a linear dispersion, which is a hallmark originating from the dipolar coupling according to Eq. (1). Note that the data are only consistent with a dipolar wave number $q_{D,\text{exp}} = 0.006 \text{ \AA} \approx q_{D,\text{lit}}/2$, i.e., smaller than reported previously [52]. Similarly, Kindervater *et al.* reported a dipolar wave number being temperature dependent and smaller in the vicinity of T_C than expected based on their measurements of the critical fluctuations in Fe [18].

B. Dynamical scaling

While dipolar interactions leave behind their signature in the spin-wave dispersion, reports of their influence on the critical scaling behavior in ferromagnets are scarce, especially in itinerant magnets. To verify the applicability of dynamical scaling theory in Ni, the linewidths determined for various $T \geq T_C$ are divided by the linewidth at T_C . This procedure reveals directly the dynamical scaling function $\gamma(x) = \Gamma(T)/\Gamma(T_C)$ (see Sec. II C). For a purely isotropic Heisenberg ferromagnet, all data points should collapse onto a universal curve, with the entire temperature dependence absorbed in the dynamical scaling variable $x = \kappa(t)/q$. Again, κ and t are the inverse correlation length and reduced temperature, respectively.

Figure 5 shows the remarkably good agreement between $\gamma(x)$ as derived from the MIEZE data and the theoretical predictions of Résibois and Piette [28] and Iro [29], which are drawn as a solid and a dashed line, respectively. Both calculations are based on the model of the isotropic

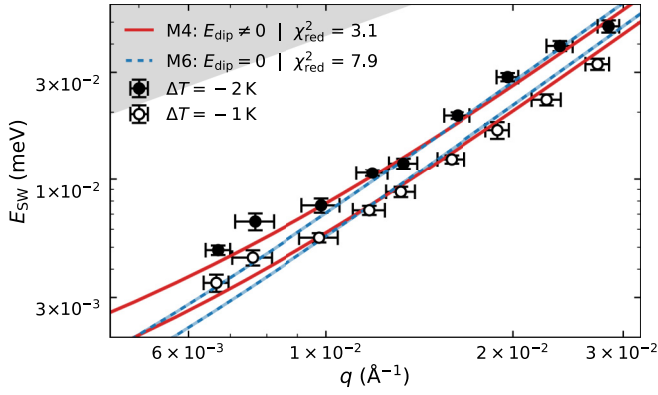


FIG. 4. Spin-wave dispersion of nickel extracted from the MIEZE data for $T = T_C - 1$ K and $T = T_C - 2$ K. The dispersion data are shown in a log-log representation together with models M4 and M6, which differ by either including (red line) or excluding (blue broken line) the dipolar interactions, respectively. The model with $E_{\text{dip}} \neq 0$ achieves a significantly lower χ^2_{red} value, therefore strongly indicating the significance of the dipolar coupling in Ni. The gray shaded area indicates the region that is inaccessible for inelastic neutron scattering due to constraints from momentum and energy conservation.

Heisenberg ferromagnet but are performed in the framework of mode coupling and asymptotic renormalization-group theory, respectively. The scaling functions have been calculated using the approximations suggested by Glinka *et al.* [30] and Iro [48].

Mode coupling approximates the data better for $\kappa/q < 1$, while the asymptotic renormalization group fits slightly better at larger κ/q . On the basis of the available data, it is unreasonable to give preference to one theory or the other. The data up to $T_C + 4$ K ($1.0063T_C$) indisputably exhibit dynamical scaling behavior expected for an isotropic Heisenberg ferromagnet.

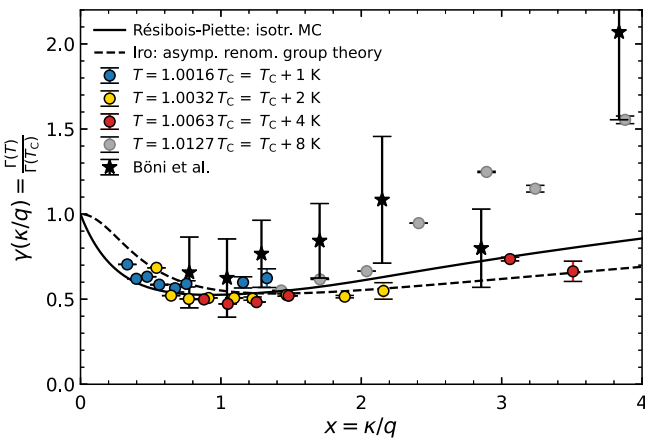


FIG. 5. Dynamic scaling function measured in nickel. The linewidth ratio $\Gamma_{T+}(q)/\Gamma_{T_C}(q)$ is supposed to lie on a universal curve described by $\gamma(\kappa/q)$. Results acquired for $\Delta T = 1$ K, 2 K, 4 K satisfy this expectation well, while for $\Delta T = 8$ K, the deviation for large $\kappa/q > 2$ is significant. Comparing the findings with TAS data from Böni *et al.* [21] shows good agreement for $\kappa/q \approx 1$. For more details see the text in Sec. IV B.

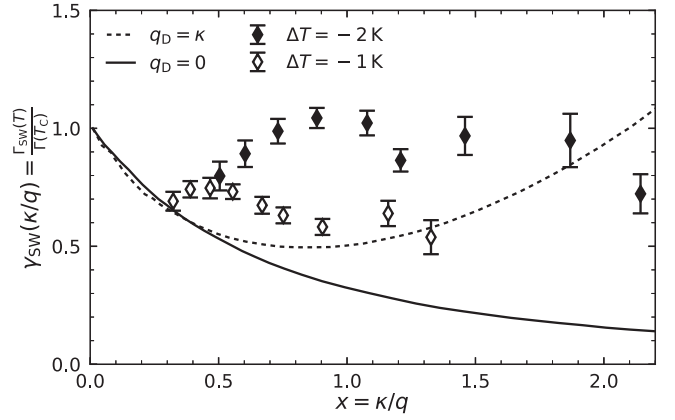


FIG. 6. Dynamical scaling function of the linewidth of the measured spin waves in Ni below T_C . Using mode-coupling theory, Schinz and Schwabl [53] calculated the dynamical scaling function of the transverse excitation modes of a dipolar ferromagnet. Two examples, representing the isotropic Heisenberg ferromagnet (black solid line) and a dipolar ferromagnet with $q_D = \kappa$ (broken line) have been added. See the text for a more detailed explanation.

Next we briefly comment on the deviation of the theoretical $\gamma(x)$ from the data for $T_C + 8$ K ($1.0127T_C$). For this purpose we have included in Fig. 5 the only other dataset available from Ni, measured by Böni *et al.* [21] using triple-axis spectroscopy (TAS). Their data were taken at significantly higher temperatures ($1.05T_C$, $1.10T_C$, $1.23T_C$, $1.48T_C$) and q values (0.009 , 0.012 Å^{-1}). Qualitatively, the data agree with the trend of the present data for $\Delta T = 8$ K ($1.0127T_C$). For a more detailed discussion of this observation see the next section.

The investigation of the spin-wave dispersion has only been made possible due to the renormalization of the spin-wave energy close to T_C , which moves their energies into the energy range observable with the MIEZE setup. Due to the excellent energy resolution, this investigation revealed a strong dependence of the linewidth on the wave vector transfer due to the renormalization. The increase of the linewidth results from the reduced spin-wave lifetime. Again, the recorded linewidth data can readily be compared with the predictions of dynamical scaling theory, which can be written as $\Gamma_{\text{SW}}(q) = \Lambda \gamma_{\text{SW}}(\kappa/q) q^z$, $z = 5/2$ in the limit of small q .

The linewidth data for $T < T_C$ are shown in Fig. 6 using a representation of the data in terms of the dynamical scaling function similar to that in Fig. 5. To reveal the scaling function $\gamma_{\text{SW}}(\kappa/q)$ directly, we divided the linewidth of the spin-wave excitations by the linewidth at T_C . In contrast to the data above T_C the data do not collapse onto a universal curve solely depending on the scaling variable κ/q . The deviations between the two datasets are clearly outside the error limits that only take into account statistical deviations. A contribution that is not captured by the error bars is the fact that there is a difference of the average wave vector transfer q between $T = T_C$ and $T < T_C$. However, this effect alone also cannot explain the deviations. We also note that close to T_C , E_{SW} and γ_{SW} are strongly correlated.

In a comprehensive theoretical treatment of the dynamics of dipolar ferromagnets below T_C , Schinz and Schwabl

derived mode-coupling equations and solved for the dynamical scaling functions numerically [53]. Their results for the isotropic case $q_D = 0$ (black solid line) and the special case of $q_D = \kappa$ (black broken line) are included in Fig. 6. As the original curve in Schinz and Schwabl [53] is parametrized as a function of $R = (\kappa^2 + q_D^2)^{1/2}/q$ the abscissa has been rescaled by a factor of $\sqrt{2}$ to be correctly represented here. However, neither the isotropic case ($q_D = 0$) nor the dipolar case ($q_D = \kappa$) of the mode-coupling results qualitatively reproduces the experimentally observed scaling behavior. Importantly, the discrepancy of the monotonous decrease of γ_{SW} in the isotropic case is striking in comparison to the data, which remain approximately constant. In contrast, the dipolar case rejects a behavior $\gamma_{SW} \rightarrow 0$ for $x \gg 0$, which is more consistent with the data within the investigated region of x . It is notable that the comparison with the dipolar case should only illustrate the general trend, but still affirms an influence of dipolar interactions in the scaling behavior below T_C . Due to the enormous complexity of the numerical calculations, we have not attempted to reproduce them for the precise parameters of our experiment. This will be discussed in more detail below.

V. DISCUSSION

The MIEZE results of nickel leave two questions open for debate: (i) Why do the measurements at $T_C + 8$ K differ from the otherwise consistent scaling behavior, and can the reasoning be extended to the data of Böni *et al.*? (ii) Why is there no indication of dipolar coupling in the dynamical scaling behavior above T_C , which arguably influences the spin waves below T_C , and has been observed in other ferromagnetic systems such as EuS [54], EuO [10,32], and Fe [18,55]?

Regarding (i), the MIEZE data at $\Delta T = 8$ K suffer from the low magnetic scattering intensity introducing systematic uncertainties not captured by the error bars in Fig. 5. The analysis algorithm implies the fitting of a sinusoidal intensity distribution in time to raw data that does not clearly exhibit sinusoidal behavior. Since the signal from the critical scattering above T_C drops sharply with increasing T , the statistics and quality of the raw data are the limiting factors, and a closer look at the reliability of the data is required to understand the behavior fully. The experimental data for $\gamma(x)$ at $\Delta T = 8$ K are shown mostly for completeness.

The same reasoning cannot be applied for the data published by Böni *et al.* because the background signal is well known and the deconvolution of the TAS data can be conducted reliably. Thus, coincidences with the MIEZE data need to be considered fortuitous. For the TAS data recorded at the two highest temperatures, one may speculate that electron-hole excitations may contribute to an additional damping of paramagnetic fluctuations. The smearing of the Fermi statistics increases the number of accessible electron states close to the Fermi surface, which can be excited by decaying, magnetic fluctuations. The details of these decay channels depend on the electronic band structure, which is not universal. This effect was also held responsible for the observed temperature dependence of the dipolar wave number q_D in Fe [18]. Generally, it is not known to what extent the band structure in metallic ferromagnets influences the properties like the

dimensionality, the range of interactions, and the conservation of the order parameter that are important for dynamical scaling theory.

While the exact conditions at which the dynamical scaling behavior in Ni breaks down are unknown, studies of critical scattering in Co [30] and Fe [56] show that for $\kappa/q > 4$ the characteristic frequency reflects the expected quadratic relationship $\omega_c(q) = A_h q^2$ with the spin-diffusion constant A_h . Hence it is conceivable that the data by Böni *et al.* and possibly the MIEZE data at $\Delta T = 8$ K begin to display an intermediate behavior dubbed "quasihydrodynamic" by Parette and Kahn [56]. It is indisputable that further research would be needed to clarify this hypothesis.

Addressing question (ii), the apparent absence of dipolar interactions in the scaling behavior of the magnetic dynamics in Ni above T_C can be understood in comparison with the results of Böni *et al.* on EuS [54] and calculations by Frey *et al.* [57]. Their studies indicate that in the paramagnetic regime, the crossover from exchange dominated to dipolar dominated dynamics only occurs deep within the dipolar critical region for fluctuations with transverse polarization, i.e., for $q, \kappa \ll q_D$. In the framework of mode-coupling theory, Frey *et al.* estimated the appearance of dipolar effects at T_C for $q/q_D \leq 0.1$ for these modes, which is consistent with investigations on EuS and Fe. Given the small value of q_D in Ni, the regime of dipolar scaling above T_C lies outside the probed parameter range.

The scaling behavior below T_C is more involved and has not been studied comprehensively. Extending the mode-coupling framework to below T_C , Schinz and Schwabl [53] considered the persistence of the magnetization, which introduces a dependence of the scaling function γ_{SW} on the orientation of $\mathbf{q}$ with respect to the magnetization $\mathbf{M}$. While their calculation reproduces and complements the understanding of the dispersion of the spin waves as they generalize the result of Holstein and Primakoff [24], the scaling function γ_{SW} fails to reproduce the experimentally observed scaling behavior (Fig. 6). Similarly, Böni *et al.* observed a constant dynamic scaling function of critical fluctuations for $T < T_C$ in Ni [20], but the data are limited to temperatures further below T_C and our study extends the observation to larger κ/q .

Based on our results and analysis process, we have identified three points for improvement that could be addressed in future studies in order to gain an even deeper understanding of the critical dynamics in ferromagnets below T_C . The first point relates to the precise control of the experimental setting. The finite magnetization $\mathbf{M}$ establishes a preferential direction and, in conjunction with $\mathbf{q}$, an intrinsic coordinate system that governs the magnetic dynamics. In our experiment, these conditions were not well controlled as high sample temperature, minuscule magnetocrystalline anisotropy, and sample environment with small stray fields allow for a random distribution of ferromagnetic domains below T_C to occur. For a more appropriate comparison of our data with theory, we would need to account for the random distribution and calculate a "powder average" of the scaling function. Preparing a magnetized Ni crystal and carefully approaching T_C from below could result in a cleaner dataset selectively resolving the components of the generalized magnetic susceptibility tensor.

The second point addresses the extraction of the linewidth parameters Γ_{SW} , which is not model agnostic and assumed two Lorentzians as the spectral weight function [see Eq. (A1)]. The resulting fit to the contrast data exhibits slight, systematic deviations at larger q , indicating a change of the spectral weight function. Here, numerical results or analytical approximations could have lead to a more accurate determination of the linewidth, which has proven to be crucial in the processing of the data for temperatures $T > T_C$ as well. However, a reimplementation of the code calculating the line shape function from mode-coupling theory [53] was found to be not feasible and therefore denying the improvement of this point, within the scope of our project.

Lastly, the third point is concerned with the computation of fine-tuned $\gamma_{\text{SW}}(x)$ curves. In Fig. 6, we compare our data to the best fitting, only available theoretical result including dipolar coupling. The curve describes a scenario of $\kappa = q_D$ (compare with Fig. 3 in Schinz and Schwabl [53]), which does not reflect the exact parameters q_D , κ , and θ realized in the measurement. θ is the angle between $\mathbf{M}$ and $\mathbf{q}$. While a dedicated calculation of $\gamma_{\text{SW}}(x)$ might achieve slightly better agreement, it is unlikely that the central prediction of the theory, namely, the minimum of the dynamical scaling function near $R/\sqrt{2} \approx \kappa/q = 1$, will change drastically. However, this predicted signature is not validated by our data and such a discrepancy also appears in the results of Böni *et al.* on EuS [54]. Given the complexity of solving the mode-coupling equations numerically, again, we cannot attempt to resolve this discrepancy ourselves.

VI. CONCLUSION

In the present work, we have determined the spin dynamics in Ni close to T_C down to very small wave vectors $q_{\text{min}} = 6.6 \times 10^{-3} \text{ \AA}^{-1}$, which is significantly smaller than the wave vectors probed so far. In addition, we used the MIEZE technique to achieve a high-energy resolution of $\Delta E = 4.9 \text{ \mu eV}$, an energy resolution that has rarely been attained in any Ni experiments.

Firstly, we succeeded to determine the dipolar wave number from the analysis of the measured spin-wave dispersion close to T_C to be around $q_D = 0.007 \pm 0.001 \text{ \AA}^{-1}$. The uncertainty is estimated only on the basis of propagating statistical uncertainties of the analysis. This value is approximately a factor of 2 smaller than the estimated value derived by Kötzler [52], i.e., $q_D = 0.013 \text{ \AA}^{-1}$. As expected, we did not observe dipolar effects on the damping of the paramagnetic fluctuations above T_C , because the probed q values are still too large. For example, in EuS, dipolar effects on the transverse fluctuations start to appear roughly at $q < q_D/5$ [31]. Therefore, to observe them in Ni, q would have to be smaller than $q \simeq 0.0014 \text{ \AA}^{-1}$, which is experimentally challenging.

Secondly, we demonstrated good agreement of the spin-wave stiffness $D(T)$ with previous work and uncovered evidence that the dynamical scaling behavior of the spin-wave linewidth $\gamma_{\text{SW}}(\kappa/q)$ is consistent with dynamical scaling of a dipolar ferromagnet.

Thirdly, we have shown that the linewidth of the critical fluctuations in Ni slightly above T_C shows scaling behavior in agreement with the scaling function derived by Résibois and

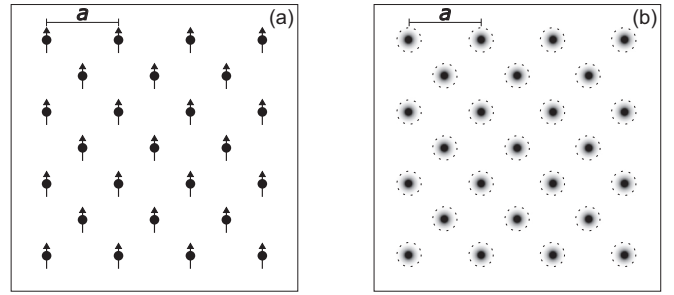


FIG. 7. (a) Visualization of the spin lattice of a Heisenberg ferromagnet. The bar labeled with a represents the lattice constant. Panel (b) shows a sketch of the real-space magnetic moment density $\rho(r)$ map of the magnetically active $3d$ electrons in Ni as determined by Mook [25]. The shading in gray corresponds to the therein published MMD values. The dashed circles represent $\rho(r) \simeq 0$. Furthermore, the experimental observation of strong localization is in good agreement with the band structure calculations of Hodges *et al.* [61].

Piette [28] and renormalization-group theory by Iro [48]. This result is remarkable because Ni represents a typical itinerant ferromagnetic system, while the scaling function is based on a Heisenberg model with short-range interactions, where the dynamical critical exponent is $z \simeq 2.5$ [Eq. (6)]. In a fully itinerant model, an exponent $z_{\text{itin}} = 3$ is expected [58,59].

Given this result a strong localization of the electrons, which contribute to the magnetic scattering, can be anticipated. This is well confirmed by experimental studies of the magnetic moment density (MMD) in metallic Ni by means of polarized neutron scattering [25] (Fig. 7). Such experiments have shown that the magnetic moment density responsible for magnetic neutron scattering is strongly localized at the atomic sites with the MMD decaying to zero after $\frac{1}{4}$ the distance to the nearest neighboring atom [25,60]. These results could be successfully understood and verified by band structure calculations, e.g., based on a parametrized Hubbard-Kanamori Hamiltonian especially suitable for the treatment of d -electron wave functions [61,62].

Concluding, in order to fully understand the effects of the dipolar interactions on the spin dynamics in itinerant electron systems, the MIEZE technique should be extended to the q regime of small-angle neutron scattering (SANS), which seems to be feasible if the two RF-spin flippers and a polarizer are installed in the collimating section of a SANS instrument. This way, it will become feasible to measure dynamical scaling functions in itinerant systems such as Ni, Ni₃Al, or MnSi even in the dipolar regime.

Moreover, it would be interesting to measure the dynamical scaling function up to high temperatures in order to identify the influence of electron-hole excitations on the damping of the paramagnetic fluctuations to bridge the gap between the MIEZE and the TAS measurements discussed above.

ACKNOWLEDGMENTS

Thanks go to Pablo Ruiz Cuevas, who supported the development and implementation of an adaptive integration algorithm to calculate the MIEZE contrast. Further acknowledgment goes to M. Antic for his great support in setting up

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DATA AVAILABILITY

The raw data acquired at RESEDA, the Python scripts that created the figures, and the additional resources are made available via Zenodo [63]. Support can be provided upon reasonable request.

APPENDIX A: DYNAMICAL SCALING THEORY AND LINE SHAPE FUNCTIONS

This section aims at summarizing the equations describing the line shape functions used for the extraction of parameters (Sec. III C) and for determining the dynamical scaling function $\gamma(x)$ (Sec. IV B), which have not been described in the text in sufficient detail.

1. Spin-wave region ($T < T_C$)

The process for extracting the linewidth and energy of the spin-wave excitations from the measured contrast is essentially the same as the procedures applied for standard neutron spin-echo data. Most commonly, an inelastic scattering process from spin waves is modeled as two Lorentzian spectral weight functions centered at a finite energy transfer $E_{\text{SW}}(q) = E_q$ with a linewidth $\Gamma_{\text{SW}}(q) = \Gamma_q$ (half width at half maximum):

$$\mathcal{L}(E|E_q, \Gamma_q) = \frac{1}{2\pi} \left(\frac{\Gamma_q}{\Gamma_q^2 + (E - E_q)^2} + \frac{\Gamma_q}{\Gamma_q^2 + (E + E_q)^2} \right). \quad (\text{A1})$$

2. Critical scaling and hydrodynamic region ($T > T_C$)

In the regime, $T > T_C$, the dynamical shape function derived by Iro using renormalization-group theory is given by

$$f_{q\xi}(x_1, s) = 2 \operatorname{Re} \left\{ \frac{1}{-is + [Z(x_1)\Pi_1(x_1, is \frac{Z(x_1)}{X(x_1)})]^{-1}} \right\}, \quad (\text{A2})$$

where, $s = E/E_c$, Π_1 is the self-energy of the dynamic susceptibility [29], and $X(x_1) = (1 + 1/x_1^2)^{-1}$ is the scaled magnetic susceptibility defined by $\chi(q, \xi) = q^{-2}X(q\xi)$ such that $x_1 = q\xi = x^{-1}$. Note that in Iro's paper the scaling variable x_1 is defined as the inverse compared to x in Sec. II C of the present work. $Z(x_1)$ is an implicit function involved in the calculation of the characteristic frequency ω_c (linewidth) of the system, which in general can only be solved numerically. However, Iro proposed the following analytical approximation for $Z(x)$:

$$Z(x_1) = \left[1 - k \arctan \left(a \frac{1 + 1/x_1^2}{(1 + b/x_1^2)^2} \right) \right]^{-1} \left(1 + \frac{b}{x_1^2} \right)^{-\epsilon/4}, \quad (\text{A3})$$

where $a = 0.46$, $b = 3.16$, and $k = 0.51$ are numerical parameters without direct physical significance. a is determined by a

calculation of the relaxation rate performed by Bhattacharjee and Ferrell [64]. b is chosen such that the dynamical scaling function $\gamma(x_1)$ asymptotically fits the behavior of the scaling function of Résibois and Piette [28]. Subsequently, k takes the value to best approximate $Z(x_1)$. $\epsilon = 6 - d = 3$ refers to the dimensionality of the renormalization group, where $d = 3$ is the dimensionality of space.

Iro also extracted the dynamical scaling function $\gamma(x_1)$ from his derivation of the magnetization correlation function. The scaling function is given by

$$\gamma(x_1) = \frac{Z(x_1)}{X(x_1)Z(\infty)}. \quad (\text{A4})$$

While the numerical values of the constants a , b , and k have been chosen with reasonable assumptions in mind, e.g., asymptotic behavior for $x_1 \rightarrow 0$, Iro argues that they can be used as fit parameters, when the theory is compared with experimental data (see, for example, Böni *et al.* [21]). For the complete derivation of the dynamical shape function the reader is referred to the original paper of Iro [29], and references therein.

Earlier, Résibois & Piette derived the scaling function $\gamma(x)$, with $x = \kappa/q$, for a Heisenberg ferromagnet [28] from mode-coupling equations. Again, these equations can only be solved numerically. Glinka *et al.* [30] approximated the numerical solution with the heuristic equation

$$\gamma(x) = e^{-1.83x} + 0.43x^{1/2}. \quad (\text{A5})$$

This parametrization was used in Fig. 5.

APPENDIX B: MODELING OF THE SPIN-WAVE DISPERSION WITH DIPOLAR INTERACTIONS

We conducted a detailed analysis of the dispersion in Ni using the Holstein-Primakoff theory and its temperature dependence as predicted by dynamical scaling theory, using several models that vary in how they take into account the contribution of the dipolar energy to the dispersion. Six models, referred to as MX ($X = 1, 2, \dots, 6$), were fit to the data, with D' as a fit parameter and varying treatments of q_D :

M1: The dipolar wave number was taken from the literature, $q_D = 0.013 \text{ \AA}^{-1}$ [52], and the contribution to the dispersion was calculated according to Eq. (4).

M2: q_D and D' were treated as free fit parameters, describing the dipolar energy as $E_{\text{dip}} = D(T)q_D^2$.

M3: q_D was determined according to the power law in Eq. (2) and $E_{\text{dip}} = g\mu_0\mu_B M(T) \sin^2(\theta_q)$. M' was set to the value of the saturation magnetization $M_{\text{sat}} = 4.91 \times 10^5 \text{ A m}^{-1}$ [65].

M4: q_D was determined from relative magnetization data $M(T)/M_{\text{sat}}$ measured by Neugebauer [51] and again $M_{\text{sat}} = 4.91 \times 10^5 \text{ A m}^{-1}$.

M5: $q_D(\Delta T = -1 \text{ K})$, $q_D(\Delta T = -2 \text{ K})$, and D' were fitted independently.

M6: Dipolar interactions were neglected and a simple quadratic dispersion was used: $E_{\text{SW}} = D(T)q^2$.

Each model has been fitted to the data fixing the critical exponent μ (and β if contained in the model) to either

TABLE I. Parameters obtained by fitting the data using the Holstein-Primakoff theory. Each model has been evaluated with $\mu = 1/3$ and 0.3407 separately. Within one cell, the upper and lower q_D values correspond to $\Delta T = -1$ K and $\Delta T = -2$ K, respectively. In models M2 and M5, q_D was a fit parameter and its uncertainty could be estimated directly from the covariance matrix. In the models M3 and M4, the uncertainty Δq_D arises from $\Delta D'$ and was calculated via Gaussian error propagation. In models M1 and M6, q_D was fixed at 0.013 and 0.0 $\text{\AA}^{-1}$, respectively.

Model	D' (meV $\text{\AA}^2$)	q_D ($10^{-3} \text{\AA}^{-1}$)	μ	χ^2_{red}
M1	333.2 ± 8.7	13.0	0.3333	4.9
	348.6 ± 8.0	13.0	0.3407	4.6
M2	382.8 ± 20.8	9.02 ± 1.50	0.3333	3.7
	400.6 ± 21.3	9.02 ± 1.47	0.3407	3.5
M3	343.5 ± 10.9	12.29 ± 0.20	0.3333	4.4
	378.1 ± 10.4	10.75 ± 0.15	0.3407	3.5
M4	412.1 ± 9.7	5.70 ± 0.07	0.3333	3.2
	431.1 ± 10.0	5.71 ± 0.07	0.3407	3.1
M5	381.6 ± 16.3	7.28 ± 0.81	0.3333	2.3
	399.5 ± 16.9	7.33 ± 0.81	0.3407	2.2
M6	454.4 ± 15.6	0.0	0.3333	8.1
	475.4 ± 16.1	0.0	0.3407	7.9

the mean-field values or the renormalization-group values. A summary of all fit parameters, the estimates of their uncertainty, and χ^2_{red} are given in Table I and Fig. 8. We find that regardless of the model, a larger μ leads to a smaller χ^2_{red} value, indicating that the critical exponents as obtained by field-theoretical methods describe the data marginally better than the mean-field values. Comparing the different models, we can see that model M5 achieves the best overall χ^2_{red} value, and therefore the best agreement with the data. Considering that M5 is the model with the fewest constraints on its fit parameters—as $q_D(\Delta T = -1 \text{ K})$, $q_D(\Delta T = -2 \text{ K})$, and D' are fit independently—this is very plausible. We find that q_D for $\Delta T = -1 \text{ K}$ is smaller than for $\Delta T = -2 \text{ K}$ and both are smaller than the literature value $q_D = 0.013 \text{\AA}^{-1}$.

Model M4, the only one based on experimental magnetization data, shows good agreement with the data based on its χ^2_{red} value. It performs better than M3, where q_D is determined by utilizing the power law of the magnetization as predicted by scaling theory. The values of q_D in model M4 are a factor of 2 smaller than in M3 and the relative difference between $q_D(\Delta T = -2 \text{ K})$ and $q_D(\Delta T = -1 \text{ K})$ is larger. This is due to the fact that the experimental magnetization decreases slower than the power law with $\beta = 0.3647$. For $\beta = 1/3$ the tem-

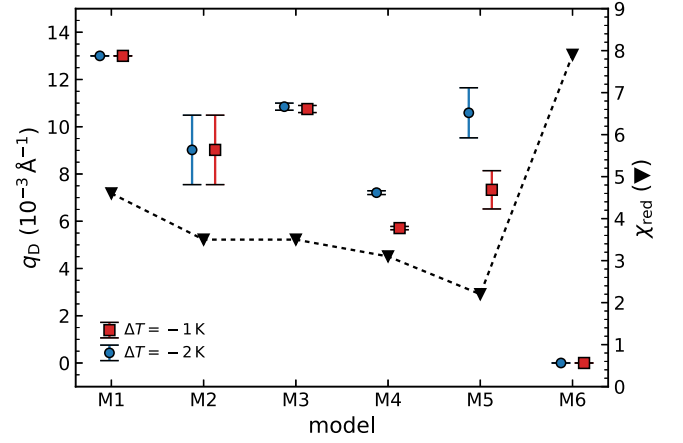


FIG. 8. Overview of the performance of Holstein-Primakoff models M1–M6 employing various strategies to include the dipolar wave number. The plot depicts $q_D(\Delta T)$ and χ^2_{red} for M1–M6 using the critical exponent $\mu = 0.3407$ as a fixed value. The data suggest $q_D \approx 0.007 \text{\AA}^{-1}$, which is half of the literature value $0.013 \text{\AA}^{-1}$, but simultaneously rejects model M6 that neglects the dipolar interactions.

perature dependence of q_D disappears completely for M3. As a consequence, D' is significantly smaller for M3 since the dipolar contribution is considerably larger. Additionally, the penalty to the χ^2 function is larger if the low- q data are fitted insufficiently. Experimental data support the lower q_D value, used for M4.

Model M1 fixes q_D at its literature value $0.013 \text{\AA}^{-1}$ and performs similarly as M3. Comparing M1 and M2, M2 achieves better agreement with $q_D(\text{M1}) > q_D(\text{M2})$.

Model M6 neglects q_D entirely, returning D' values that are slightly larger than those of the other models. The deviation from the dipolar behavior can be best seen at low q , as evidenced when looking at the first few data points for each temperature in Fig. 4. This discrepancy shows that the inclusion of dipolar interactions is in fact required.

In the literature, the influence of the dipolar coupling on the critical dynamics mostly focused on the temperature regime above T_C , with a limited number of published experiments directly addressing the spin-wave regime [14,19] below T_C . Especially Ni has been disregarded in the past as the combination of a large spin-wave stiffness and a small dipolar wave number makes observations of q_D experimentally challenging. Nevertheless, measuring the spin-wave energy below q_D should show a clear imprint leading to an approximately linear dispersion $E_{\text{SW}} \approx D(T)q_D q$. However, a linear contribution to the dispersion is not easily visible in Fig. 4, even though the lowest q value is a factor of 2 smaller than the reported $q_D = 0.013 \text{\AA}^{-1}$ [52].

It is apparent that the measured dispersion is consistent with $0.006 \text{\AA}^{-1} < q_D < 0.011 \text{\AA}^{-1}$. Comparing the performance of M4 and M5, it is reasonable to assume a significant temperature dependence of q_D , which has already been pointed out by Böni *et al.* [14]. However, this dependence is more pronounced than expected from dynamical scaling theory with $q_D^2 \propto M/D \propto |t|^{\beta-\mu}$ ($\beta - \mu = 0.024$) for a 3D Heisenberg system [50]. Since only two different tempera-

tures could be measured, a reliable analysis of the temperature dependence is impossible and requires a more detailed in-

vestigation over a wider T range and by extending the measurements to smaller q .

- [1] R. D. Lowde and C. G. Windsor, Generalized susceptibility of nickel, *Phys. Rev. Lett.* **18**, 1136 (1967).
- [2] V. J. Minkiewicz, M. F. Collins, R. Nathans, and G. Shirane, Critical and spin-wave fluctuations in nickel by neutron scattering, *Phys. Rev.* **182**, 624 (1969).
- [3] H. A. Mook, J. W. Lynn, and R. M. Nicklow, Temperature dependence of the magnetic excitations in nickel, *Phys. Rev. Lett.* **30**, 556 (1973).
- [4] J. W. Lynn and H. A. Mook, Temperature dependence of the dynamic susceptibility of nickel, *Phys. Rev. B* **23**, 198 (1981).
- [5] Y. J. Uemura, G. Shirane, O. Steinsvoll, and J. Wicksted, New explanation of the spin-wave-like excitations in Ni above T_c , *Phys. Rev. Lett.* **51**, 2322 (1983).
- [6] O. Steinsvoll, C. F. Majkrzak, G. Shirane, and J. Wicksted, Paramagnetic scattering from metallic Ni, *Phys. Rev. B* **30**, 2377 (1984).
- [7] H. A. Mook, D. M. Paul, and S. Hayden, Temperature dependence of the high-energy magnetic excitations for Ni, *Phys. Rev. B* **38**, 12058(R) (1988).
- [8] G. Shirane, Magnetic correlations in 3d ferromagnets above T_c , *J. Magn. Magn. Mater.* **45**, 33 (1984).
- [9] Y. Endoh and P. Böni, Magnetic excitations in metallic ferro- and antiferromagnets, *J. Phys. Soc. Jpn.* **75**, 111002 (2006).
- [10] O. W. Dietrich, J. Als-Nielsen, and L. Passell, Neutron scattering from the Heisenberg ferromagnets EuO and EuS. III. Spin dynamics of EuO, *Phys. Rev. B* **14**, 4923 (1976).
- [11] J. Als-Nielsen, O. W. Dietrich, and L. Passell, Neutron scattering from the Heisenberg ferromagnets EuO and EuS. II. Static critical properties, *Phys. Rev. B* **14**, 4908 (1976).
- [12] P. Böni and G. Shirane, Paramagnetic neutron scattering from the Heisenberg ferromagnet EuO, *Phys. Rev. B* **33**, 3012 (1986).
- [13] S. Schorr, P. Vorderwisch, and F. Mezei, Critical dynamics in EuO below the Curie point, *Physica B* **234-236**, 749 (1997).
- [14] P. Böni, M. Hennion, and J. L. Martínez, Magnon dispersion of the dipolar ferromagnet EuS near the zone center, *Phys. Rev. B* **52**, 10142 (1995).
- [15] G. Shirane, Y. J. Uemura, J. P. Wicksted, Y. Endoh, and Y. Ishikawa, Paramagnetic scattering from a cubic ferromagnet Pd_2MnSn , *Phys. Rev. B* **31**, 1227 (1985).
- [16] H. A. Graf, P. Böni, G. Shirane, M. Kohgi, and Y. Endoh, Paramagnetic neutron scattering from Pd_2MnSn , *Phys. Rev. B* **40**, 4243 (1989).
- [17] F. Mezei, Role of spin-nonconserving forces in the critical dynamics of Fe at the Curie point, *Phys. Rev. Lett.* **49**, 1096 (1982).
- [18] J. Kindervater, S. Säubert, and P. Böni, Dipolar effects on the critical fluctuations in Fe: Investigation by the neutron spin-echo technique MIEZE, *Phys. Rev. B* **95**, 014429 (2017).
- [19] S. Säubert, J. Kindervater, F. Haslbeck, C. Franz, M. Skoulatos, and P. Böni, Dipolar interactions in Fe: A study with the neutron Larmor precession technique MIEZE in a longitudinal field configuration, *Phys. Rev. B* **99**, 184423 (2019).
- [20] P. Böni, J. L. Martínez, and J. M. Tranquada, Longitudinal spin fluctuations in nickel, *Phys. Rev. B* **43**, 575 (1991).
- [21] P. Böni, H. A. Mook, J. L. Martínez, and G. Shirane, Comparison of the paramagnetic spin fluctuations in nickel with asymptotic renormalization-group theory, *Phys. Rev. B* **47**, 3171 (1993).
- [22] Y. Ishikawa, Y. Noda, C. Fincher, and G. Shirane, Low-energy paramagnetic spin fluctuations in the weak itinerant ferromagnet mnsi, *Phys. Rev. B* **25**, 254 (1982).
- [23] B. I. Halperin and P. C. Hohenberg, Generalization of scaling laws to dynamical properties of a system near its critical point, *Phys. Rev. Lett.* **19**, 700 (1967).
- [24] T. Holstein and H. Primakoff, Field dependence of the intrinsic domain magnetization of a ferromagnet, *Phys. Rev.* **58**, 1098 (1940).
- [25] H. A. Mook, Magnetic moment distribution of nickel metal, *Phys. Rev.* **148**, 495 (1966).
- [26] B. I. Halperin and P. C. Hohenberg, Scaling laws for dynamic critical phenomena, *Phys. Rev.* **177**, 952 (1969).
- [27] R. Folk and H. Iro, Paramagnetic neutron scattering and renormalization-group theory for isotropic ferromagnets at T_c , *Phys. Rev. B* **32**, 1880 (1985).
- [28] P. Résibois and C. Piette, Temperature dependence of the linewidth in critical spin fluctuation, *Phys. Rev. Lett.* **24**, 514 (1970).
- [29] H. Iro, The dynamical correlation function for isotropic ferromagnets for $T \geq T_c$, *Z. Phys. B* **68**, 485 (1987).
- [30] C. J. Glinka, V. J. Minkiewicz, and L. Passell, Small-angle critical neutron scattering from cobalt, *Phys. Rev. B* **16**, 4084 (1977).
- [31] P. Böni, D. Görlitz, J. Kötzler, and J. L. Martínez, Dynamics of longitudinal and transverse fluctuations above T_c in EuS, *Phys. Rev. B* **43**, 8755 (1991).
- [32] F. Mezei, Critical dynamics in EuO at the ferromagnetic Curie point, *Physica B+C* **136**, 417 (1986).
- [33] E. Frey and F. Schwabl, Critical dynamics of dipolar ferromagnets, *Phys. Lett. A* **123**, 49 (1987).
- [34] E. Frey and F. Schwabl, On the critical dynamics of ferromagnets, *Z. Phys. B* **71**, 355 (1988).
- [35] E. Frey and F. Schwabl, Critical dynamics of magnets, *Adv. Phys.* **43**, 577 (1994).
- [36] F. Mezei, Neutron spin echo: A new concept in polarized thermal neutron techniques, *Z. Phys.* **255**, 146 (1972).
- [37] R. Gähler, R. Golub, and T. Keller, Neutron resonance spin echo—a new tool for high resolution spectroscopy, *Physica B* **180-181**, 899 (1992).
- [38] C. Franz, O. Soltwedel, C. Fuchs, S. Säubert, F. Haslbeck, A. Wendl, J.K. Jochum, P. Böni, and C. Pfeleiderer, The longitudinal neutron resonant spin echo spectrometer RESEDA, *Nucl. Instrum. Methods Phys. Res., Sect. A* **939**, 22 (2019).
- [39] R. Gähler, R. Golub, K. Habicht, T. Keller, and J. Felber, Space-time description of neutron spin echo spectrometry, *Physica B* **229**, 1 (1996).
- [40] F. Mezei, C. Pappas, and T. Gutberlet, *Neutron Spin Echo Spectroscopy: Basics, Trends and Applications* (Springer Science & Business Media, Berlin, 2002), Vol. 601.

- [41] L. Van Hove, Correlations in space and time and born approximation scattering in systems of interacting particles, *Phys. Rev.* **95**, 249 (1954).
- [42] C. Franz, O. Soltwedel, S. Säubert, A. Wendl, W. Gottwald, F. Haslbeck, L. Spitz, and C. Pfeiderer, Longitudinal neutron resonance spin echo spectroscopy under large energy transfers, *J. Phys.: Conf. Ser.* **1316**, 012005 (2019).
- [43] L. Beddrich, Consequences of lattice dimensionality in solid state magnetism, Ph.D. thesis, Technische Universität München, 2023.
- [44] L. Beddrich, J. K. Jochum, P. Bender, L. Spitz, A. Wendl, C. Franz, S. Busch, F. Juranyi, C. Pfeiderer, and O. Soltwedel, Comparison of time-of-flight and MIEZE neutron spectroscopy of H₂O, *J. Appl. Crystallogr.* **58**, 1122 (2025).
- [45] M. Köhli, M. Klein, F. Allmendinger, A.-K. Perrevoort, T. Schröder, N. Martin, C. J. Schmidt, and U. Schmidt, Cascade—a multi-layer boron-10 neutron detection system, *J. Phys.: Conf. Ser.* **746**, 012003 (2016).
- [46] Heinz Maier-Leibnitz Zentrum (MLZ), Experimental facilities Heinz Maier-Leibnitz Zentrum (MLZ) (2022), https://www.frm2.tum.de/fileadmin/w00bnv/www/content/uploads/experimental-facilities_online.pdf.
- [47] A. Schober, A. Wendl, F. X. Haslbeck, J. K. Jochum, L. Spitz, and C. Franz, The software package MIEZEPY for the reduction of MIEZE data, *J. Phys. Commun.* **3**, 103001 (2019).
- [48] H. Iro, The dynamical correlation function of an isotropic heisenberg ferromagnet in the critical region, *J. Magn. Magn. Mater.* **73**, 175 (1988).
- [49] B. Hammouda and D. F. R. Mildner, Small-angle neutron scattering resolution with refractive optics, *J. Appl. Crystallogr.* **40**, 250 (2007).
- [50] J. C. Le Guillou and J. Zinn-Justin, Critical exponents for the n -vector model in three dimensions from field theory, *Phys. Rev. Lett.* **39**, 95 (1977).
- [51] C. A. Neugebauer, Temperature dependence of the saturation magnetization of nickel films of thickness less than 100Å, *J. Appl. Phys.* **31**, S152 (1960).
- [52] J. Kötzler, Critical phenomena in dipolar magnets, *J. Magn. Magn. Mater.* **54-57**, 649 (1986).
- [53] H. Schinz and F. Schwabl, Dynamics of dipolar ferromagnets below T_c , *Phys. Rev. B* **57**, 8438 (1998).
- [54] P. Böni, B. Roessli, D. Görlitz, and J. Kötzler, Damping of spin waves and singularity of the longitudinal modes in the dipolar critical regime of the heisenberg ferromagnet EuS, *Phys. Rev. B* **65**, 144434 (2002).
- [55] F. Mezei, Critical dynamics in isotropic ferromagnets, *J. Magn. Magn. Mater.* **45**, 67 (1984).
- [56] G. Parette and R. Kahn, Étude de la diffusion critique des neutrons par le fer dans les régions « hydrodynamique » et « quasi hydrodynamique », *J. Phys. France* **32**, 447 (1971).
- [57] E. Frey, F. Schwabl, and S. Thoma, Shape functions of dipolar ferromagnets at and above the curie point, *Phys. Rev. B* **40**, 7199 (1989).
- [58] P. Böni and G. Shirane, Paramagnetic neutron scattering from ni (invited), *J. Appl. Phys.* **57**, 3012 (1985).
- [59] G. G. Lonzarich and L. Taillefer, Effect of spin fluctuations on the magnetic equation of state of ferromagnetic or nearly ferromagnetic metals, *J. Phys. C: Solid State Phys.* **18**, 4339 (1985).
- [60] L. Dobrzynski, R. J. Papoular, and M. Sakata, Internal magnetization density distribution of iron and nickel by the maximum entropy method, *J. Phys. Soc. Jpn.* **65**, 255 (1996).
- [61] L. Hodges, N. D. Lang, H. Ehrenreich, and A. J. Freeman, Magnetic form factor of nickel, *J. Appl. Phys.* **37**, 1449 (1966).
- [62] P.-W. Ma and S. L. Dudarev, Nonuniversal structure of point defects in face-centered cubic metals, *Phys. Rev. Mater.* **5**, 013601 (2021).
- [63] L. Beddrich, J. K. Jochum, C. Franz, S. Säubert, and P. Böni, Supplementary material - The influence of dipolar interactions on the critical dynamics in nickel, Zenodo (2026), <https://doi.org/10.5281/zenodo.17347394>.
- [64] J. K. Bhattacharjee and R. A. Ferrell, Dynamic scaling for the isotropic ferromagnet: ϵ expansion to two-loop order, *Phys. Rev. B* **24**, 6480 (1981).
- [65] J. Crangle, The magnetization of pure iron and nickel, *Proc. R. Soc. London, Ser. A* **321**, 15 (1971).