

Nd(Co_{0.8}Fe_{0.2})₂化合物在居里转变处的临界指数研究

翟琳¹, 郑文根²

¹南京信息职业技术学院电子信息学院, 江苏 南京

²南京航空航天大学物理学院, 江苏 南京

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摘要

本文通过直流磁化测量研究了Nd(Co_{0.8}Fe_{0.2})₂在居里温度(T_C)附近的临界行为。利用Kouvel-Fisher法、改进的Arrotts图法以及临界等温分析, 获得了临界指数 $\beta = 0.38(5)$ 、 $\gamma = 1.31(4)$ 和 $\delta = 4.41(3)$ 。通过 $M|t|^{-\beta}$ 与 $H|t|^{-(\beta+\gamma)}$ 的对数标度图验证了所得临界指数的可靠性。此外, 临界指数 β 、 γ 、 δ 以及交换相互作用 $J(r) \sim 1/r^{4.87}$ 表明, Nd(Co_{0.8}Fe_{0.2})₂符合三维海森堡模型, 其中短程相互作用占主导地位。

关键词

相变, 临界行为

Critical Exponents at Curie Transition in Nd(Co_{0.8}Fe_{0.2})₂ Compound

Lin Zhai¹, Wengen Zheng²

¹School of Electronic Information, Nanjing Vocational College of Information Technology, Nanjing Jiangsu

²Department of Applied Physics, Nanjing University of Aeronautics and Astronautics, Nanjing Jiangsu

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Abstract

The critical behavior around the Curie temperature (T_C) of Nd(Co_{0.8}Fe_{0.2})₂ was investigated by dc magnetization measurement. The critical exponents $\beta = 0.38(5)$, $\gamma = 1.31(4)$ and $\delta = 4.41(3)$

were carried out by Kouvel-Fisher, modified Arrotts Plot methods, as well as critical isothermal analysis. The reliability of the obtained critical exponents was verified by the logarithmic scaling plot of $M|t|^{-\beta}$ versus $H|t|^{-(\beta+\gamma)}$. Besides, the exponents β , γ , and δ , as well as the exchange interaction $J(r) \sim 1/r^{4.87}$ proposed a three-dimensional Heisenberg model for $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ in which the short-range interaction becomes dominant.

Keywords

Phase Transitions, Critical Behavior

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1. 引言

近几十年来, 由于在磁制冷技术中的潜在应用价值, 研发具有优异磁制冷性能材料一直是凝聚态物理领域的重点研究方向[1]-[3]。近年来, 具有丰富的磁学特性及相对简单的晶体结构的 RT_2 型 Laves 相化合物(其中 R 为稀土金属, T 为过渡金属)成为磁学材料领域的研究热点[4]-[7]。在众多材料体系中, RCo_2 基化合物因其显著的磁热效应(MCE)而备受关注[8]-[11]。例如, DyCo_2 、 HoCo_2 、 ErCo_2 等化合物因其在居里温度 T_c 附近具有较大的磁熵变而成为磁热效应研究中最具应用潜力的候选材料[6] [9] [12] [13]。然而, 一级相变材料通常存在显著的热滞与磁滞现象, 且工作温区相对狭窄, 这成为制约其实际应用的关键瓶颈。近年来, 研究发现赝二元化合物 $\text{Nd}(\text{Co}_{1-x}\text{Fe}_x)_2$ 展现出巨磁熵变与宽工作温区的特性, 这为磁制冷技术带来重要突破[14] [15]。在 $\text{Nd}(\text{Co}_{1-x}\text{Fe}_x)_2$ 中掺杂 Fe 元素可提升居里温度(T_c)并拓宽工作温区, 该规律同样适用于 $\text{A}(\text{Co}_{1-x}\text{Fe}_x)_2$ 体系($\text{A} = \text{Dy}, \text{Tb}, \text{Er}, \text{Ho}$) [16]-[20]。但是, 这一现象背后的物理机制至今仍未完全阐明。要理解 $\text{Nd}(\text{Co}_{1-x}\text{Fe}_x)_2$ 的磁性物理性质, 明确化合物中磁相互作用的类型至关重要。在磁性体系相变研究中, 临界指数(β , γ , δ)能够有效表征化合物的相变本质。其中有三种经典模型: 三维海森堡模型($\beta = 0.365$, $\gamma = 1.386$)、三维伊辛模型($\beta = 0.325$, $\gamma = 1.241$)和平均场理论模型($\beta = 0.5$, $\gamma = 1$), 而前两者描述短程磁相互作用, 后者表征长程相互作用[21]。

本文针对 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 合金在顺磁-铁磁相变边界处的临界行为展开了系统研究, 通过修正 Arrott 图技术(MAP) [22]与 Kouvel-Fisher (KF)方法[23], 实验测得临界指数。结果通过 $M|t|^{-\beta}$ 与 $H|t|^{-(\beta+\gamma)}$ 的修正等温线得以验证。我们的研究证实, $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 磁相变附近的临界行为符合具有短程铁磁相互作用的三维海森堡模型。

2. 实验方法

单相的 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 化合物通过高纯度电弧熔炼及后续退火工艺制备。实验中添加 5%过量 Nd 以补偿烧损并防止富 Co 相的形成。样品为单一的 Laves 相结构[11] [15]。磁化等温线测量在 250 至 280 K 的升温过程中、最高 30 kOe 外磁场下完成。

3. 结果与讨论

先前工作显示 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 化合物的居里温度为 263 K [15]。因此, 我们在 263 K 附近(250~280 K)

以 2 K 为步长、磁场从 0 至 30 kOe 递增的条件下测量了等温磁化强度, 如图 1(a)所示。

在相变理论中, 临界指数通常通过以下理论模型给出:

$$M_s(T) = M_0 |t|^\beta \quad (t < 0) \quad (1)$$

$$\chi_0^{-1}(T) = \left(\frac{h_0}{M_0} \right) t^\gamma \quad (t > 0) \quad (2)$$

$$M = DH^{1/\delta} \quad (t = 0) \quad (3)$$

其中 $t = (T - T_c)/T_c$, M_0 , h_0/M_0 和 D 为临界振幅, $M_s(T)$ 是自发磁化强度, $\chi_0^{-1}(T)$ 代表温度为 T 时初始磁化率的倒数[24][25]。假设 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 的磁化强度服从平均场模型($\beta = 0.5$, $\gamma = 1$), 其 M^2 与 H/M 的关系曲线如图 1(b)所示。根据 Banerjee 判据, $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 的磁相变被确认为二级相变, 其核心实验证据为 M^2 vs. H/M 曲线的斜率为正[25]。此外, 根据 Arrott-Noakes 公式[22],

$$(H/M)^{1/\gamma} = at + bM^{1/\beta} \quad (4)$$

其中 a 和 b 为常数, 不同温度下的 $M^{1/\beta}$ 对 $(H/M)^{1/\gamma}$ 曲线(如图 1(b)所示)在高场范围内应呈平行关系。同时, 居里温度 T_c 对应的直线应通过原点。因此, 图 1(b)中不平行的曲线表明平均场模型不适用。

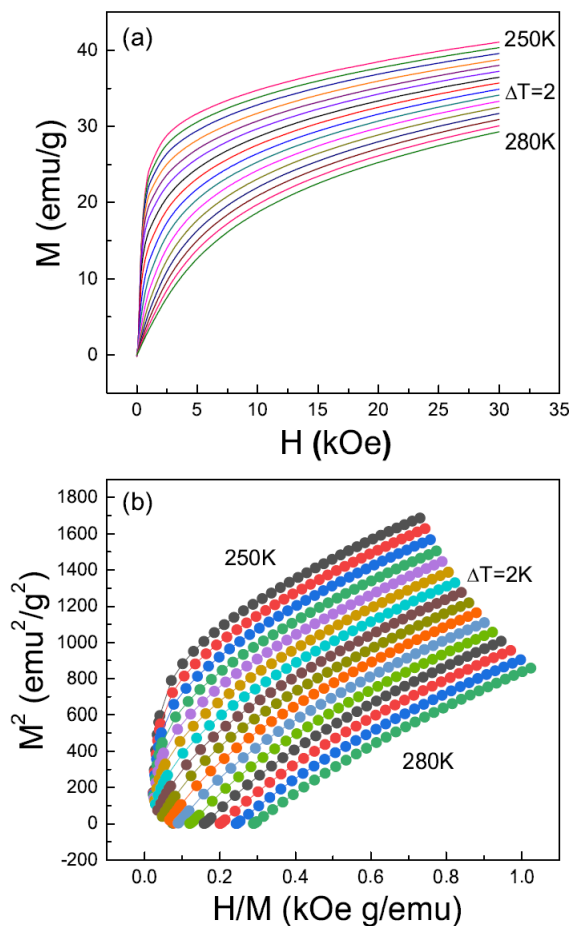


Figure 1. (a) (Color online) (a) Isothermal magnetization curves of $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ near T_c . (b) Arrott plot isotherms of M^2 versus H/M curves

图 1. (a) (在线彩图) $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 在居里温度(T_c)附近的等温磁化曲线 (b) M^2 对 H/M 的 Arrott 等温线图

为进一步获得可靠的 β 和 γ , 我们使用了改进的 Arrott 图(MAP)方法[22]。以初始值 $\beta=0.5$ 、 $\gamma=1$ 为基础, 通过高场区线性外推与 $M^{1/\beta}$ 轴和 $(H/M)^{1/\gamma}$ 轴的交点, 分别获得居里温度以下的饱和磁化强度 $M_s(T)$ 和居里温度以上的初始磁化率倒数 $\chi_0^{-1}(T)$ 。通过将 $M_s(T)$ 和 $\chi_0^{-1}(T)$ 分别与方程(1)和(2)拟合, 可获得新的 β 和 γ 值。随后, 这些 β 和 γ 值将作为新的初始值用于下一轮计算。经过几轮迭代后, 结果收敛于 $\beta=0.38(5)$ 和 $\gamma=1.31(4)$ 。采用 $\beta=0.38(5)$ 和 $\gamma=1.31(4)$ 绘制的新 MAP 图展示在图 2 中。高场下平行的等温线以及几乎呈线性且过原点的 264 K 曲线证实了 β 和 γ 的可靠性。低场下线性的偏移可以解释为磁畴的重新排列[26]。饱和磁化强度 M_s 和初始磁化率倒数 χ_0^{-1} 的温度依赖性及 $\beta=0.38(5)$ 、 $\gamma=1.31(4)$ 的拟合结果如图 3(a)所示。两条拟合曲线分别给出居里温度为 262.34 (9) K 和 262.36 (8) K。

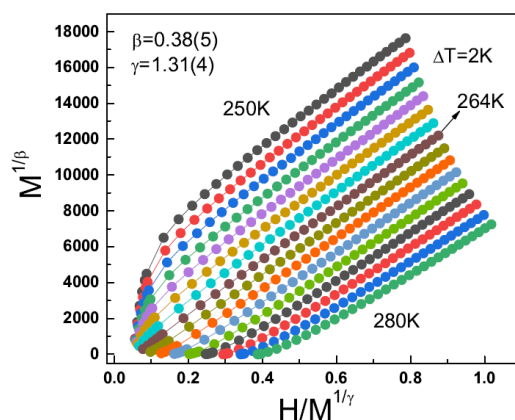


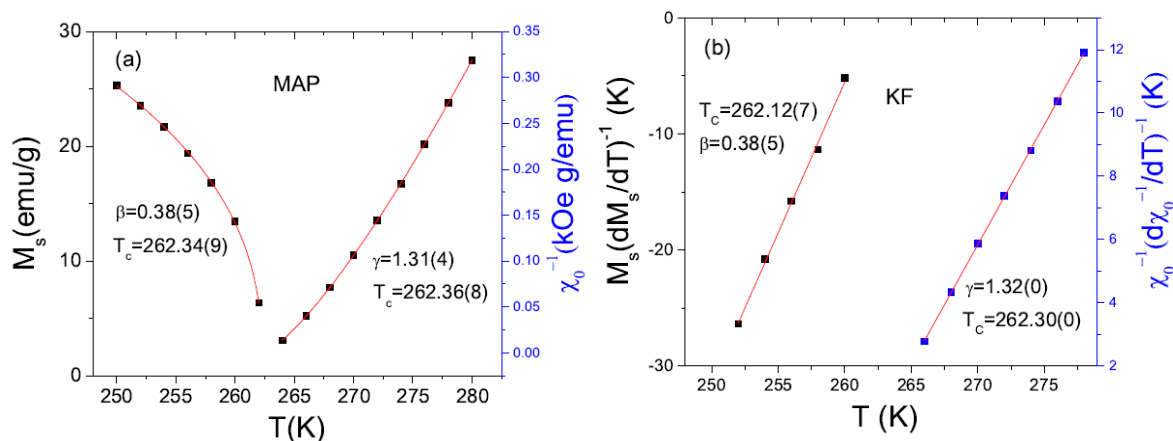
Figure 2. (Color online) Modified Arrott plot isotherms of M^2 versus H/M with the critical exponents $\beta=0.38(5)$ and $\gamma=1.31(4)$

图 2. (在线彩图)修正 Arrott 图: M^2 对 H/M 的等温线图(含临界指数 $\beta=0.38(5)$ 和 $\gamma=1.31(4)$)

同时, KF 方法也是一种计算临界指数的方法, 其相关公式为[23]:

$$\frac{M_s(T)}{dM_s(T)/dT} = \frac{T-T_C}{\beta} \quad (5)$$

$$\frac{\chi_0^{-1}(T)}{d\chi_0^{-1}(T)/dT} = \frac{T-T_C}{\gamma} \quad (6)$$



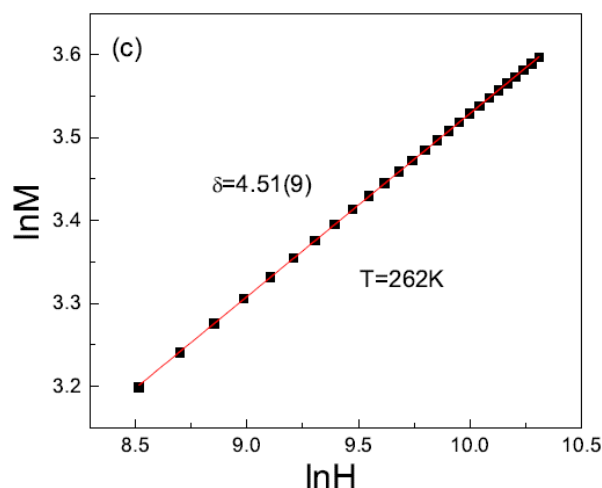


Figure 3. (Color online) (a) and (b) The fits of M_s and χ_0^{-1} using MAP methods and KF methods, respectively. (c) $\ln M$ versus $\ln H$ curve at 262 K, $\delta = 4.51(9)$ is obtained by the linear fit (red line)

图 3. (在线彩图) (a)和(b)分别利用 MAP 和 KF 方法对饱和磁化强度 $M_s(T)$ 和初始磁化率倒数 $\chi_0^{-1}(T)$ 的拟合. (c) 262 K 下的 $\ln M$ vs. $\ln H$ 曲线及临界指数 δ

因此, 通过 $\frac{M_s(T)}{dM_s(T)/dT}$ 和 $\frac{\chi_0^{-1}(T)}{d\chi_0^{-1}(T)/dT}$ 对温度 T 的斜率可以分别得到 β 和 γ 。如图 3(b)所示, 利用式(5)拟合得到 $\beta = 0.38(5)$ 且居里温度 $T_c = 262.17(7)$, 而式(6)给出 $\gamma = 1.32(0)$ 且 $T_c = 262.30(0)$ 。MAP 方法与 KF 方法的结果相互吻合, 且接近三维海森堡模型预测的值 ($\beta = 0.368$, $\gamma = 1.396$) [27]。图 3(c)展示了在居里温度 T_c 下 $\ln H$ 对 $\ln M$ 的曲线, 根据式(3), 斜率的倒数给出 $\delta = 4.51(9)$ 。另一方面, Widom 标度关系也是另一种计算 δ 值的方法[28]。

$$\delta = 1 + \frac{\gamma}{\beta} \quad (7)$$

采用 MAP 方法和 KF 方法分别获得 $\delta = 4.41(3)$ 和 $\delta = 4.42(9)$, 二者在实验误差范围内均与标度假设一致。

此外, 通过标度分析验证了所得临界指数的可靠性。根据标度假设, 实验测得的 $M|t|^{-\beta}$ 与 $H|t|^{-(\beta+\gamma)}$ 满足以下关系:

$$M|t|^{-\beta} = f_{\pm}(H|t|^{-(\beta+\gamma)}) \quad (8)$$

对于 $T > T_c$ 和 $T < T_c$, f_+ 和 f_- 分别为正则函数[29]。因此, 实验测得的 $M(H)$ 曲线在 $T > T_c$ 和 $T < T_c$ 时应塌缩为两条普适曲线。图 4 展示了利用 $\beta = 0.38(5)$ 和 $\gamma = 1.31(4)$ 绘制的 $M|t|^{-\beta}$ 与 $H|t|^{-(\beta+\gamma)}$ 对数标度图, 两个独立分支证实了 β 和 γ 的可靠性。

此外, $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 中的自旋相互作用可通过关系式 $J(r) \sim 1/r^{d+\sigma}$ 估算, 其中 d 为空间维度, σ 为相互作用范围参数, 该参数可由方程求得[30] [31]。

$$\gamma = 1 + \frac{4}{d} \left[\frac{n+2}{n+8} \right] \Delta\sigma + \frac{8(n+2)(n+4)}{d^2(n+8)^2} \times \left[1 + \frac{2G\left(\frac{d}{2}\right)(7n+20)}{(n-4)(n+8)} \right] (\Delta\sigma)^2 \quad (9)$$

其中 $\Delta\sigma = \left(\sigma - \frac{d}{2}\right)$ 且 $G\left(\frac{d}{2}\right) = 3 - \frac{1}{4}\left(\frac{d}{2}\right)^2$ 。由于 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 被归类为三维海森堡模型, 因此采用空间维度 $d=3$ 和自旋维度 $n=3$ 。根据 MAP 方法获得的 $\gamma=1.31(4)$, 计算得到 $\sigma=1.87(3)$ 。因此, 交换相互作用表现为 $J(r) \sim 1/r^{4.87}$, 这与具有短程磁相互作用的三维海森堡模型 $J(r) \sim 1/r^5$ 相当。另一方面, NdCo_2 中居里温度以下长程磁有序的存在已通过粉末中子衍射证实[32]。 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 中从长程到短程磁相互作用的转变可能是由于掺杂 Fe 导致化合物中磁矩的局域化引起的[33]。

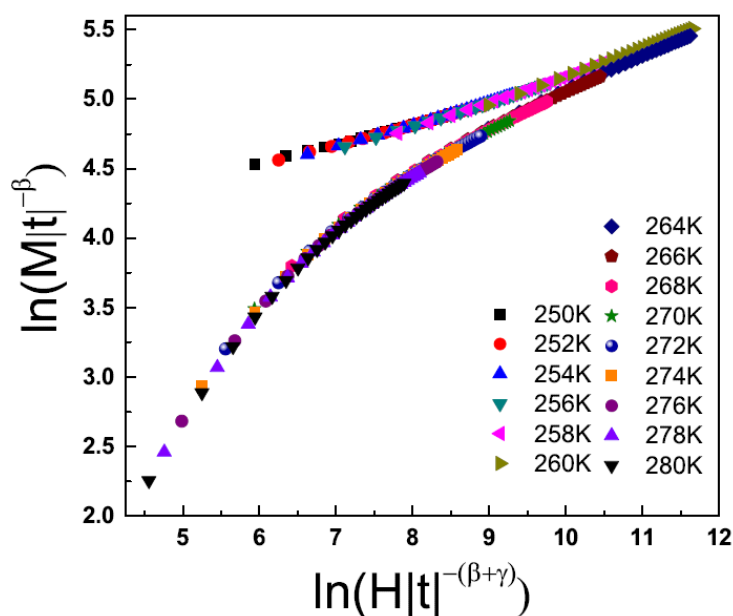


Figure 4. (Color online) Logarithmic scaling plot of $M|t|^{-\beta}$ versus $H|t|^{-(\beta+\gamma)}$ with $\beta=0.38(5)$ and $\gamma=1.31(4)$ for $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$

图 4. (在线彩图): $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 的 $M|t|^{-\beta}$ 与 $H|t|^{-(\beta+\gamma)}$ 对数标度图

4. 结论

我们对 $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 在居里温度附近的临界行为进行了研究。Banerjee 判据和普适行为均表明该体系发生二级相变, 且实验测得的相变温度与计算值一致。采用改进的 Arrott 图(MAP)方法和 KF 方法对临界特性进行了研究, 两种方法结果吻合。临界指数值及自旋相互作用研究表明, $\text{Nd}(\text{Co}_{0.8}\text{Fe}_{0.2})_2$ 中存在服从三维海森堡模型的短程磁相互作用。

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