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Properties of $GdAlO_3$ near the Spin-Flop Bicritical Point

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I have determined the phase boundaries, magnetization discontinuity across the spin-flop transition, and divergence of the susceptibility close to the bicritical point in antiferromagnetic $GdAlO_3$. The experiments yield the first confirmation of the predictions of renormalization-group studies of the anisotropy crossover exponent, orientation of scaling axes, and some exponent relations.

In a uniaxial antiferromagnet, the paramagnetic (PM) phase orders into an antiferromagnetic (AF) phase in zero and low fields, but into a spin-flop (FL) phase in high fields. For the applied field H along the easy axis, a first-order spin-flop (SF) transition separates the longitudinally ordered AF phase from the perpendicularly ordered FL phase and joins the PM phase boundaries $T_c^{\parallel}(H)$ and $T_c^{\perp}(H)$ in a bicritical point T_b (Fig. 1). Fisher and Nelson[1] have shown that the competition of the two orderings in the vicinity of the bicritical point gives rise to a new type of critical behavior governed by the anisotropy crossover exponent, ϕ. I have experimentally tested their predictions on how the two PM boundary branches meet at T_b, the way the magnetization discontinuity across the SF transition vanishes, and the power with which the direct susceptibility diverges at T_b. These experiments allow one, for the first time, to obtain ϕ in a direct way and, using the universal critical-line amplitude ratio calculated in the preceding Letter,[2] to verify the predicted orientation for the magnetic field scaling axis.[2]

The experiments have been carried out on a bar of $GdAlO_3$ of 0.5 mm×0.5 mm×12 mm, with the easy axis along the long axis of the bar, cut out of a large single crystal grown from flux by the accelerated crucible-rotation technique.[3] $GdAlO_3$ has orthorhombic anisotropy[4] and its critical properties are those of an $n=2$ component Heisenberg system.[1] Experimentally, a small misalignment of the applied field with respect to the easy axis cannot be avoided. Such a misalignment, however, changes the phase diagram near T_b in a crucial way. As shown pre-

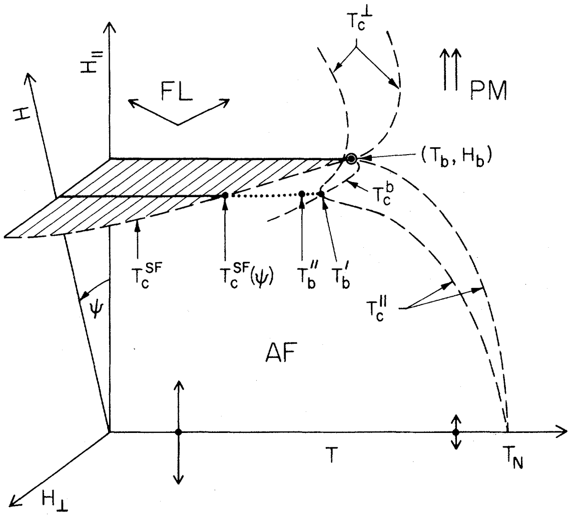

FIG. 1. Schematic phase boundaries of a uniaxial antiferromagnet near the bicritical point T_b. The PM-AF ($T_c^{\parallel}$) and the PM-FL ($T_c^{\perp}$) critical lines are shown for perfect alignment ($H_{\parallel}$) and for misalignment ψ; the pseudobicritical point T_b' lies on the critical line $T_c^{\,b}$. The shaded surface of the first-order spin-flop transition is bounded by the spin-flop critical line $T_c^{\,\mathrm{SF}}$. The dotted line between $T_c^{\,\mathrm{SF}}(\psi)$ and T_b' indicates the pseudo SF transition line explained in the text.

viously,[5,6] the SF transition is confined to a narrow triangular shelf perpendicular to the applied field (see Fig. 1). Thus for finite misalignment angle ψ the SF transition becomes disconnected from the PM boundary and ends at a critical point $T_c^{\,\mathrm{SF}}(\psi)$ on the spin-flop critical line $T_c^{\,\mathrm{SF}}$. In the transitionless region between $T_c^{\,\mathrm{SF}}(\psi)$ and the PM boundary, the system changes gradually in increasing field, in a narrow field interval, from an AF-like to an FL-like configuration. This gives rise to a peak in the susceptibility $\tilde{\chi}(H)$ rather than a magnetization discontinuity. The locus of these peaks is a smooth continuation of the SF transition line. We call it the pseudo SF transition line and its intersection with the PM boundary locates the pseudobicritical point T_b'. The perpendicular field component at T_b' also scales like t^{ϕ} where $t = T/T_b - 1$, giving rise to a Heisenberg-like bicritical "umbilicus" at T_b.[7]

The phase diagram of Fig. 1 also applies to orthorhombic anisotropy provided the misalignment is not too close to the easy-hard plane. For small ψ, $T_c^{\,\mathrm{SF}}(\psi)$ decreases linearly with ψ—for a long cylinder of $GdAlO_3$ at a rate[6,8] $dt_c^{\,\mathrm{SF}}(\psi)/d\psi \approx 0.065\ \mathrm{deg}^{-1}$, where $t_c^{\,\mathrm{SF}}(\psi) = 1 - T_c^{\,\mathrm{SF}}(\psi)/T_b$. This is one and two orders of magnitude slower than in MnF_2 and Cr_2O_3, respectively,[5] and it makes $GdAlO_3$ a suitable material for investigations where alignment is important.

Phase boundaries.—Figure 2 shows the phase diagram determined from the maxima of the susceptibility in the vicinity of the bicritical point. The susceptibility was measured by the mutual

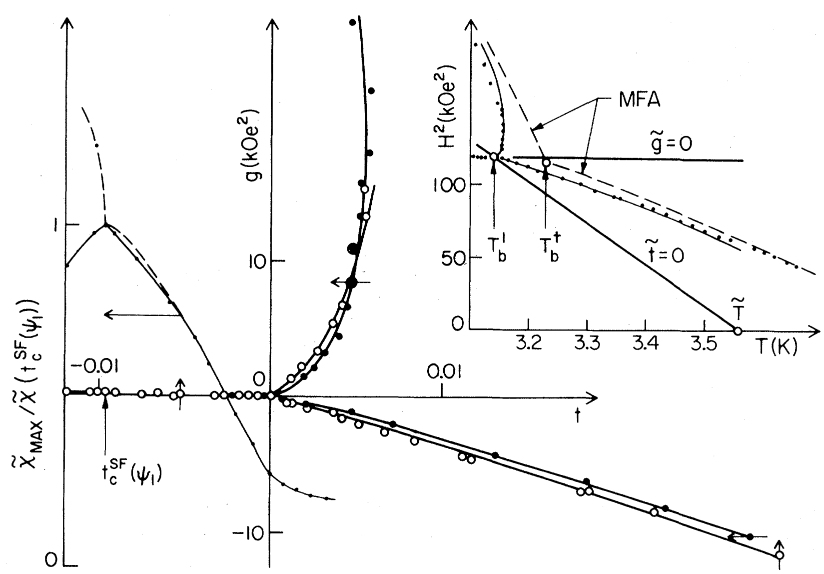

FIG. 2. Phase boundaries near the bicritical point for two misalignments $\psi_1 = 0.14°$ (open circles) and $\psi_2 \approx 0.08°$ (filled circles). The drawn PM boundaries are least-squares fits by Eq. (1). The left-hand side shows the temperature dependence of $\tilde{\chi}_{\max}(H)$ for ψ_1 along the true and pseudo SF transition. (Dashed line, isothermal; solid line, adiabatic.) The inset shows the orientation of the scaling axes, the extrapolations of the high- and low-field MFA phase boundaries, and the experimental results for ψ_2 ($T_b' = 3.141$ K, $H_b' = H_b = 10.90$ kOe).

induction method with modulation fields <10 Oe and with the pickup coil wound around the central 4 mm of the sample. Since the demagnetizing field at the bicritical point amounts to only 8 Oe, changes of demagnetization in the vicinity of the bicritical point have been neglected. Sweep rates were typically 0.1 mK/sec and 0.2 Oe/sec and the sweep directions are indicated by arrows in Fig. 2. Along the pseudo SF transition line, the susceptibility maximum $\tilde{\chi}_{max}(H)$ increases rapidly with decreasing temperature, shown on the left-hand side of Fig. 2. The onset of the true SF transition, $T_c^{SF}(\psi)$, is marked by the sharp maximum in the temperature dependence of the adiabatic susceptibility peak.[6]

Near the bicritical point, extended scaling predicts the two branches of the PM boundaries for perfect alignment to follow[1]

$$\tilde{g} = \pm w_{\perp,\parallel} \tilde{t}^{\phi}, \tag{1}$$

meeting tangetially at T_b for $\phi > 1$. The plus and minus signs go with $w_\perp$ and $w_\parallel$, respectively. The optimal scaling fields $\tilde{g}$ and $\tilde{t}$ are given by $\tilde{g} = g - pt$ and $\tilde{t} = t + gg$,[2,7] where $g = H^2 - H_b^2$ and $t = T/T_b - 1$. The $\tilde{g} = 0$ scaling axis is tangent to the SF transition line at T_b; expressions for the orientation of the $\tilde{t} = 0$ scaling axis are given in the preceding Letter.[2] For finite ψ, the PM boundaries do not meet tangentially at T_b'; however, away from T_b', they should follow Eq. (1) (see Ref. 2 and references therein) using a pseudobicritical temperature T_b'' (see Fig. 1). In the present experiment, the expected rounding at T_b' could not be resolved and thus T_b' was used for a fit to Eq. (1). In order to reduce the number of fitting parameters we make use of the scaling relation $w_\perp = w_\perp = w$.[2,7] A least-squares fit in the range $t > 0.02$ then gives $\phi = 1.15 \pm 0.08$, $q = 1.11 \times 10^{-3}$ kOe^{-2}, and $w = (1.25 \pm 0.3) \times 10^2$ kOe^2 for misalignment $\psi_1 = 0.14°$ and $\phi = 1.25 \pm 0.07$, $q = 1.12 \times 10^{-3}$ kOe^{-2}, and $w = (1.76 \pm 0.3) \times 10^3$ kOe^2 for $\psi_2 = 0.08°$, where the errors quoted correspond to a 25% increase of the mean standard deviations $\sigma_{\tilde{t}} = 0.0006$ and 0.0005, respectively.

The values thus obtained for ϕ are in reasonable agreement with the predicted $\phi = 1.175 \pm 0.015$.[1] Experimentally, the PM-FL boundary favors slightly higher values of ϕ; the PM-AF boundary, lower ones. The experiment also indicates the "umbilical" nature of the bicritical point; a quantitative analysis, however, is not possible at present. The orientation of the $\tilde{t} = 0$ scaling axis is shown in the inset of Fig. 2. Theory predicts[2]

$$\tilde{T} = T_b + (T_N - T_b^{\dagger})(n+2)/3n \tag{2}$$

(I prefer to use the alternative effective bicritical point $T_b^{\dagger}$ instead of T_b^*), where $\tilde{T}$ is the intersection of the $\tilde{t} = 0$ scaling axis with the temperature axis and $T_N = 3.878$ K[4,6] is the Néel temperature. In the spirit of the theory,[2] $T_b^{\dagger} = 3.23$ K was taken as the intersection of the two branches of the molecular field (MFA) paramagnetic boundary. These MFA boundaries are extrapolations of the high-temperature ($T > 3.68$ K) and high-field ($H > 17$ kOe) PM boundaries, where MFA behavior was observed.[6] Equation (2) gives $\tilde{T} = 3.57$ K, in excellent agreement with the experimental $\tilde{T} = 3.56$ K.

Magnetization discontinuity.—Because of the finite width of the SF transition and the large susceptibility at the pseudo SF transition, a direct determination of ΔM is difficult.[9] I have obtained ΔM by integrating the isothermal susceptibility over a narrow field interval with the corresponding area at $T_c^{SF}(\psi)$ subtracted. With $T_c^{SF}(\psi)$ determined independently of ΔM as described above, this procedure increased the useful reduced temperature range by an order of magnitude. The results are shown in Fig. 3 together with earlier measurements[10] of the width of the domain state,[11] ΔH_D, at the SF transition. Extended scaling pre-

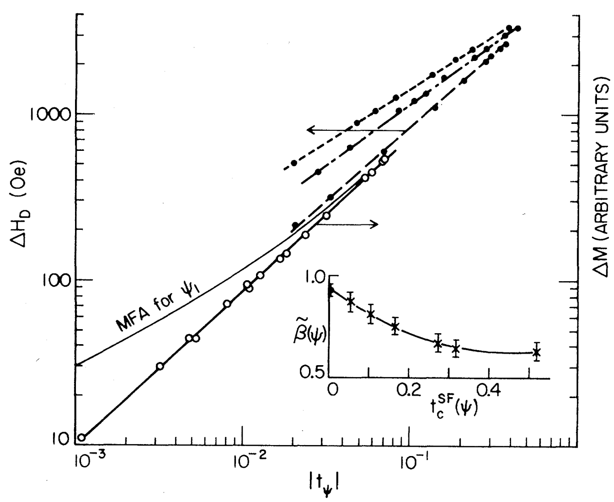

FIG. 3. Magnetization discontinuity for different misalignments. Open circles, from integrated susceptibility (this work), $t_c^{SF}(\psi_1) = 0.0096$; filled circles, from width of domain state [$t_c^{SF}(\psi) = 0.05$ ($\psi = 0.3°$), 0.16 (1°), and 0.32 (1.6°)]. Straight lines are fits by Eq. (3), curved line gives MFA for $t_c^{SF}(\psi_1) = 0.0096$ fitted to the experiment at $|t_\psi| = 0.07$. The inset shows the dependence of $\tilde{\beta}$ on misalignment.

dicts[1] for perfect alignment

$$\Delta M \propto |t|^{\tilde{\beta}}. \qquad (3)$$

The experiment shows that for finite ψ, ΔM is still of the form of Eq. (3) with $|t|$ replaced by $|t_\psi| = 1 - T/T_c^{\rm SF}(\psi)$ and $\tilde{\beta} = \tilde{\beta}(\psi)$. Extrapolation to $\psi = 0$ gives $\tilde{\beta}(0) = 0.92 \pm 0.03$ compared to the predicted $\tilde{\beta} = 0.85 \pm 0.04$. With the exponent relation[1] $\varphi = 2 - \tilde{\beta} - \alpha$, where $\alpha(n=2) = -0.02 \pm 0.02$ is the exponent of the specific heat along $\tilde{g} = 0$, one obtains $\phi = 1.10 \pm 0.05$. This is somewhat lower than the predicted value and those obtained above. Current theory does not predict $\tilde{\beta}(\psi)$ for $\psi \neq 0$, nor is the crossover behavior as ψ tends to zero understood. An extrapolation to $\psi = 0$ might therefore not be justified, as indicated by MFA where $\Delta M \propto \{|t_\psi|[|t_\psi| + t_c^{\rm SF}(\psi)T_b/T_c^{\rm SF}(\psi)]\}^{1/2}$; i.e., $\Delta M \propto |t|$ for $\psi = 0$ and $\Delta M \propto (|t_\psi|)^{1/2}$ otherwise[10] (see Fig. 3).

Susceptibility.—Along the linear scaling axis $\tilde{g} = 0$, $\tilde{\chi}$ is predicted[1] to diverge with $t^{-\tilde{\gamma}}$, where $\tilde{\gamma} = 2\varphi + \alpha - 2 = 0.33 \pm 0.05$, whereas sufficiently away from the bicritical point $\tilde{\chi}$ should reflect the considerably weaker divergence of the specific heat. We find that in the reduced temperature range 8×10^{-4} to 2.3×10^{-2}, $\tilde{\chi}(T)$ diverges with power $\tilde{\gamma}_\parallel = 0.11$ for $H \leq 10.7$ kOe, reasonably consistent with the predicted 0.125 of Ising-like behavior. For $H \geq 11.1$ kOe, $\tilde{\chi}(T)$ diverges with $\tilde{\gamma}_\perp = 0.03$ in agreement with the logarithmic divergence reported previously.[6] Closer to the bicritical point, crossover effects are observed and $\tilde{\chi}(T)$ cannot be fitted by a power law. Along $\tilde{g} = 0$, finally, a stronger divergence with $\tilde{\gamma} = 0.15$ is found, implying $\phi = 1.09$. This divergence is weaker than the predicted one; however, $\tilde{\chi}$ is expected to diverge more strongly for better alignment as indicated by the strong susceptibility increase between T_b' and $T_c^{\rm SF}(\psi)$.

In conclusion, I have experimentally verified some of the predictions for critical properties near the spin-flop bicritical point. The observed PM boundaries agree well with the predicted crossover exponent and the orientation of the scaling axes. Magnetization discontinuity and susceptibility also differ significantly from MFA but yield a somewhat smaller ϕ.

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