Magnons as a Bose-Einstein Condensate in Nanocrystalline Gadolinium

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The recent observation [S. P. Mathew *et al.*, J. Phys. Conf. Ser. **200**, 072047 (2010)] of the anomalous softening of spin-wave modes at low temperatures in nanocrystalline gadolinium is interpreted as a Bose-Einstein condensation (BEC) of magnons. A self-consistent calculation, based on the BEC picture, is shown to closely reproduce the observed temperature variations of magnetization and specific heat at constant magnetic fields.

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Bose-Einstein condensation (BEC) is a macroscopic quantum phenomenon in which, below a certain (ultralow) temperature, a macroscopic number of bosons spontaneously condense into a single lowest-energy quantum state and the spontaneous quantum coherence of BE condensate, so formed, persists over macroscopic length and time scales. A long-standing theoretical possibility that a spin system, undergoing a quantum phase transition at a critical magnetic field, H_c, where long-range magnetic order either appears or disappears, can be mapped onto a system of weakly interacting bosons and the long-range magnetic order near the quantum critical point can be modeled as BEC [1–3] has been explored extensively in the recent years. Accordingly, in spin-dimer compounds $BaCuSi_2O_6$, $Sr_3Cr_2O_8$, $Ba_3Cr_2O_8$, $ACuCl_3$ ($A =$ K, Tl, NH_4), $Pb_2V_3O_9$ [4–11], quasi-two-dimensional spin-1/2 antiferromagnet Cs_2CuCl_4 [12], and the spin-gap compound $NiCl_2$-$4SC(NH_2)_2$ [13], the phase transition from nonmagnetic (spin singlet) state to a magnetically ordered (spin triplet) state, occurring at H_c and resulting in field-induced magnetic order, has been attributed to the BEC of magnons. This interpretation has been questioned [14,15] on the basis that (i) a rearrangement of the ground state of the system by the application of field generates virtual magnons as opposed to the real ones needed for BEC and (ii) in quantum antiferromagnets, in general, and in the compounds $ACuCl_3$ ($A =$ K, Tl, NH_4), in particular, the U(1) rotational symmetry around the applied magnetic field is broken by magnetocrystalline anisotropy [16] rather than by field-induced magnetic order, as required for the BEC of gapless Goldstone magnon modes or triplons.

In a parallel development, there have been reports [17] of quasiequilibrium magnons (sustained by microwave parametric pumping in epitaxial yttrium iron garnet films) undergoing BEC at room temperature. It has, however, been argued [18] that the experiments using coherent magnon pumping demonstrate, at best, an accumulation of magnon population near the ground state but provide no direct test of spontaneous coherence, which is essential to the phenomenon of BEC. The coherence of magnon condensates witnessed in recent experiments involving incoherent pumping of magnons [19] restores some confidence in the claim of BEC.

In this Letter, we follow a completely different approach to demonstrate BEC of magnons in a new system (nanocrystalline gadolinium, nc-Gd) at temperatures $T \leq 20$ K in zero external magnetic field. In this approach, the physical quantities (that characterize the BEC transition) such as (i) the BEC transition temperature at different magnetic fields $T_c(H)$ and (ii) the average occupation number for the ground state (the BEC order parameter) $\langle n_0 \rangle$ and the chemical potential μ, as functions of temperature and magnetic field, are self-consistently determined from the magnetization and specific heat data taken on well-characterized [20,21] high-quality nc-Gd samples with average grain sizes of $d = 12$ nm and $d = 18$ nm. Previous studies on systems undergoing a magnon BEC transition yielded $T_c(H)$ but not $\langle n_0(T, H) \rangle$ and $\mu(T, H)$. This work thus marks the first attempt to experimentally determine all three quantities in any magnon BEC system.

A brief outline of the theoretical formalism used is given below. Assuming quadratic magnon dispersion at long wavelengths and measuring the wave vector (momentum) **k** from the minimum of the magnon dispersion, the low-energy effective Hamiltonian for magnons in the presence of magnetic field H is given by

$$H = \sum_k \left(\frac{\hbar^2 k^2}{2m^*} + \Delta - \mu \right) a_k^\dagger a_k + \frac{1}{2} \sum_{k,k',q} v(\mathbf{q}) a_{k+q}^\dagger a_{k'-q}^\dagger a_k a_{k'}, \quad (1)$$

where $a_k^\dagger$ and a_k are the boson creation and annihilation operators for magnons of wave vector k, the effective mass $m^* \equiv \hbar^2/2D$, D is the spin-wave (SW) stiffness, $\Delta = \Delta_0 + g\mu_B H$, Δ_0 is the gap introduced in the spin-wave spectrum by the dipole-dipole interactions and/or magnetocrystalline anisotropy while $g\mu_B H$ is the Zeeman contribution to the gap. The second term in Eq. (1) represents the four-magnon interaction and gives rise to the ‘‘remormalization’’ of D with temperature in accordance with the relation [22]

$$D(T) = D(0)[1 - D_2 T^2 - D_{5/2} T^{5/2}], \tag{2}$$

where $D(0)$ is the spin-wave stiffness at $T = 0$ K and the $T^{5/2}$ (T^2) term arises from the direct (indirect) magnon-magnon interactions (mediated by the conduction-electron spins). Considering that the Ruderman-Kittel-Kasuya-Yosida interaction is basically responsible for the ferromagnetic ground state in *nc*-Gd, the indirect magnon-magnon interactions dominate at low temperatures and hence the $T^{5/2}$ term in Eq. (2) is dropped in subsequent calculations. The momentum distribution of magnons in the normal (uncondensed) phase is

$$\bar{n}_k \equiv \langle a_k^\dagger a_k \rangle = \frac{1}{e^{\beta(\varepsilon_k + \Delta - \mu)} - 1}, \tag{3}$$

with $\varepsilon_k \equiv \hbar^2 k^2 / 2m^*$. The magnon density $n = N/V$ has to be determined self-consistently by

$$n = \sum_k \bar{n}_k = \frac{1}{(2\pi)^3} \int_0^\infty \bar{n}_k d^3k = \lambda^{-3} \sum_{l=1}^{\infty} \frac{(z_{\rm eff})^l}{l^{3/2}} \tag{4}$$

with the thermal de Broglie wavelength $\lambda = \sqrt{2\pi\hbar^2/m^* k_B T} = \sqrt{4\pi D(T)/k_B T}$, $z_{\rm eff} = z e^{-\beta\Delta}$, and the fugacity $z = e^{\beta\mu}$ given by the equation of state for non-interacting Bose gas

$$\frac{1}{V} \frac{z}{1-z} = n\left[1 - \frac{g_{3/2}(z)}{g_{3/2}(1)} \left(\frac{T}{T_c}\right)^{3/2}\right], \tag{5}$$

where V is the volume over which the condensate wave function retains its phase coherence, $\langle n_0 \rangle \equiv z/(1-z)$ is the average occupation number for the ground state, $g_{3/2}(z) \equiv \sum_{l=1}^{\infty} z^l / l^{3/2}$ is the Bose-Einstein function while the critical temperature T_c at which the thermal de Broglie wavelength becomes comparable to the average interparticle separation is

$$T_c = (2\pi\hbar^2/k_B m^*)[\zeta(3/2)]^{-2/3}(n^{2/3}). \tag{6}$$

The standard statistical mechanics treatment yields the magnetization $M(T, H)$ and the magnon contribution to specific heat per unit volume $c_{\rm mag}(T, H)$, as

$$M(T, H) = M(0, H) - g\mu_B n \tag{7}$$

with magnon density n given by Eq. (4), and

$$c_{\rm mag}(T, H) = \frac{3k_B}{2}\left(\frac{k_B T}{4\pi D(T)}\right)^{3/2} \left\{\left[\frac{5}{2} + \frac{3D_2 T^2}{1 - D_2 T^2}\right] \times \sum_{l=1}^{\infty} \frac{(z_{\rm eff})^l}{l^{5/2}} - \ln z_{\rm eff} \sum_{l=1}^{\infty} \frac{(z_{\rm eff})^l}{l^{3/2}}\right\}. \tag{8}$$

If $\mu = 0$, Eq. (7) reduces to the conventional SW relation for magnetization,

$$M(T, H) = M(0, H) - g\mu_B Z\left(\frac{3}{2}, t_H\right)\left[\frac{k_B T}{4\pi D(T)}\right]^{3/2}, \tag{9}$$

where the Bose-Einstein integral function $Z(\frac{3}{2}, t_H) = \sum_{n=1}^{\infty} n^{-3/2} \exp(-n t_H)$ with $t_H = \Delta / k_B T$. Note that the above theoretical treatment is quite general in that the same formalism or its system-specific variant can be used to describe any magnon BEC system.

Recently, an anomalous softening [21] of magnon modes at low temperatures [$T < T^*(H)$] for $H \geq 1$ kOe, where an upturn in $M(T)$ occurs, was inferred from the conventional spin-wave analysis of $M(T)$ of *nc*-Gd. By following a self-consistent approach, detailed below, we demonstrate that this softening of magnon modes is a consequence of BEC of magnons. At first, Eq. (5) is solved in conjunction with Eq. (4), for certain initial values of V and T_c, to yield fugacity as a function of temperature, $z(T)$, in a given temperature range. $z(T)$, so obtained, is inserted into Eq. (7) or (8), and $M(T, H)$ or $c_{\rm mag}(T, H)$ for a fixed H is calculated using the value $\Delta_0 = 0.155(3)$ meV, previously reported [23] for single-crystal Gd, and trial values of $D(0)$ and D_2 in Eq. (2). The calculated $f(T, H)_{\rm cal}$ is compared with the observed $f(T, H)_{\rm obs}$ (where $f \equiv M$ or the total specific heat C_H, which besides $c_{\rm mag}$ has additive electronic and phonon contributions) over the chosen temperature range and this iterative process is repeated for a different set of values for V, T_c [and hence $z(T)$], $D(0)$, and D_2 until the agreement between $f(T, H)_{\rm cal}$ and $f(T, H)_{\rm obs}$ in that temperature range is optimized. The same self-consistent procedure is followed in each temperature range as the temperature range is widened by including higher temperature data in the analysis. At a given field, $M(T, H)$ and $c_{\rm mag}(T, H)$ data yield identical values (within the uncertainty limits) for the parameters V, T_c, $D(0)$, and D_2. Figures 1 and 2 demonstrate that the BEC picture, Eqs. (7) and (8) (continuous curves), describes $M_H(T)$ and $c_{\rm mag}(T, H)$ [and hence $C_H(T)$] quite well over a temperature range which widens with H. By contrast, the conventional spin-wave theory, Eq. (9), corresponding to the $\mu = 0$ case, describes $f(T, H)_{\rm obs}$, at best, in an extremely narrow range as $T \to 0$ (inset in Fig. 1).

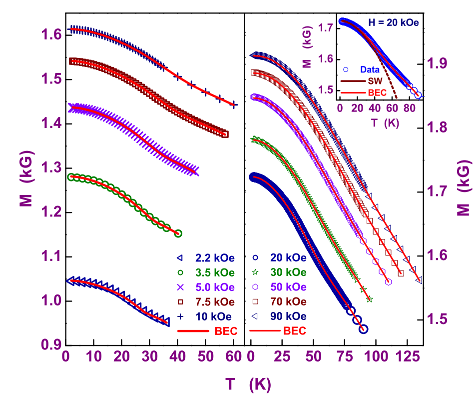

FIG. 1 (color online). Comparison between the observed (symbols) and theoretical [continuous curves, yielded by Eqs. (7), (4), and (5)] temperature variations of magnetization at fixed fields in the range 2.2–90 kOe in nanocrystalline Gd with an average grain size of 12 nm. The inset illustrates that, unlike the BEC formalism, the conventional spin-wave theory fails to describe $M(T)$ over an extended temperature range.

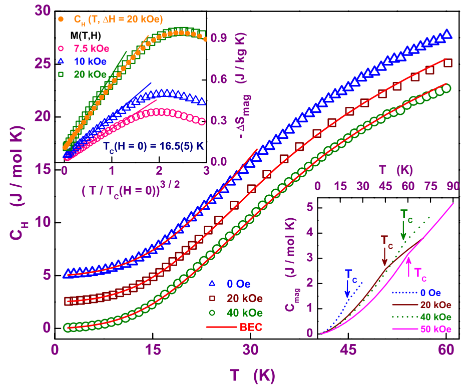

FIG. 2 (color online). The total specific heat $C_H(T)$ at a few representative fields for $d = 12$ nm. Note that the $C_{H=20\ \mathrm{kOe}}(T)$ and $C_{H=0}(T)$ data are shifted up by 2.5 and 5.0 J/mol K, respectively, with respect to that taken at $H = 40$ kOe. The continuous theoretical curves are obtained by adding the (Sommerfeld) electronic, (Debye) lattice, and (BEC) magnon C_{mag} contributions to $C_H(T)$. The bottom inset shows $C_{\mathrm{mag}}(T)$, the slope change at T_c, predicted by the BEC theory, and the increase in, and progressive smearing of, T_c with H. The top inset displays the magnetic entropy change $-\Delta S_{\mathrm{mag}}$ for a few representative ΔH values (open symbols), obtained from $M_H(T)$, plotted against $[T/T_c(H = 0)]^{3/2}$ for $d = 12$ nm. The straight lines serve to highlight the characteristic BEC $T^{3/2}$ variation of entropy for $T \ll T_c$. $-\Delta S_{\mathrm{mag}}$ at $\Delta H = 20$ kOe (solid circles), calculated from $C_H(T)$, agrees quite well with that calculated from $M_H(T)$ (open squares).

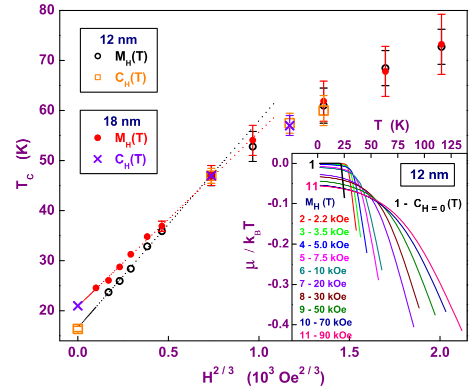

FIG. 3 (color online). The BEC transition temperature T_c obtained from $M_H(T)$ and $c_{\mathrm{mag}}(T, H)$ plotted against $H^{2/3}$ for $d = 12$ and 18 nm. The bottom inset depicts the temperature variations of the normalized chemical potential $\mu/k_B T$ at various fixed field values for $d = 12$ nm.

$c_{\mathrm{mag}}(T, H)$, shown for a few representative fields in the bottom inset of Fig. 2, exhibits the slope change at T_c that is generic to BEC. The other notable feature is the increase in, and progressive smearing of, T_c with H.

The volume V over which the condensate wave function retains its phase coherence shrinks by nearly 14 (6) orders of magnitude from its value 0.0075(5) cm^3 [$1.1(6) \times 10^{-11}$ cm^3] at $H = 0$ for $d = 12$ nm [18 nm] and approaches the volume of a single grain as fields in excess of 30 kOe are applied. The effect of magnetic field is to create a gap in the spin-wave spectrum (in addition to the intrinsic gap Δ_0), suppress spin waves, and progressively destroy phase coherence. T_c increases with H in accordance with the relation $T_c(H) = T_c(H = 0) + aH^{1/\phi}$, with the exponent $\phi = 3/2$ (Fig. 3) that is characteristic [2,8,24] of BEC, up to $H = 20$ kOe (30 kOe) for $d = 12$ nm (18 nm).

The variation of fugacity with temperature, $z(T)$, that optimizes agreement between $f(T, H)_{\mathrm{cal}}$ and $f(T, H)_{\mathrm{obs}}$ at different but fixed fields permits an accurate determination of the average occupation number for the ground state $\langle n_0 \rangle = z/(1 - z)$ and the chemical potential, $\mu = k_B T \ln z$, as functions of temperature and field. $\langle n_0(T = 1.8\ \mathrm{K}, H = 0)\rangle = 2.4(12) \times 10^{15}$ [$3.3(14) \times 10^6$] for $d = 12$ nm (18 nm). This result indicates that, in the limit $H \to 0$, a sizable fraction of the magnons [magnon density as high as $\langle n_0(T = 1.8\ \mathrm{K}, H \to 0)\rangle / V(H \to 0) \approx 3 \times 10^{17}\ \mathrm{cm}^{-3}$] excited at $T = 1.8$ K spontaneously condense into the ground state in both the samples $d = 12$ and 18 nm, but the condensate wave function retains its phase coherence over the entire sample volume only in the case of $d = 12$ nm. Incidentally, the value of intrinsic spin-wave energy gap [23] $\Delta_0 = 0.155(3)$ meV corresponds to a temperature of $T_0 = 1.80(5)$ K so that the long-wavelength magnons with a density $\sim 10^{17}$–$10^{18}\ \mathrm{cm}^{-3}$ can be easily excited at $T \geq 1.8$ K. However, regardless of the values of $\langle n_0(T = 1.8\ \mathrm{K}, H)\rangle$ and $V(H)$ at a given H (including $H = 0$) for the samples with $d = 12$ and 18 nm, the ratio $\langle n_0(T = 1.8\ \mathrm{K}, H)\rangle / V(H)$ is the same (within the uncertainty limits) for both the samples and increases with H.

In accordance with the BEC predictions, Eqs. (5) and (6), at constant H, the condensate fraction $\langle n_0(T, H)\rangle / \langle n_0(T = 1.8\ \mathrm{K}, H)\rangle$ scales with $[T/T_c(H)]^{3/2}$ over a temperature range which increases with decreasing H (Fig. 4) while $T_c(H) \propto [\langle n_0(T = 1.8\ \mathrm{K}, H)\rangle / V(H)]^{2/3}$ (inset of Fig. 4). As the coherence volume V shrinks with increasing H, the sharp kink in $z(T)$ or $\mu(T)$ at $T = T_c$ gets smeared out progressively so much so that z (or equivalently, μ) falls short of the value unity (zero) even at $T = 0$ (bottom inset of Fig. 3) with the result that the smearing of the transition at T_c occurs and the ratio $\langle n_0(T, H)\rangle / \langle n_0(T = 1.8\ \mathrm{K}, H)\rangle$ acquires higher and higher values even at temperatures in the vicinity of T_c (Fig. 4). It is clear that at temperatures well below T_c, $\mu \to 0$ only in the limit $H \to 0$. As expected for a true thermodynamic BEC phase transition, concomitant with zero chemical potential (denoted by curve 1 in the inset of Fig. 3), the condensate fraction $\langle n_0(T, H = 0)\rangle / \langle n_0(T = 1.8\ \mathrm{K}, H = 0)\rangle$, deduced from $c_{\mathrm{mag}}(T, H = 0)$, decreases linearly with the reduced temperature $[T/T_c(H = 0)]^{3/2}$ (solid line in Fig. 4) so as to drop to zero at $T = T_c(H = 0)$ and remains zero for $T \geq T_c(H = 0)$, where $\mu/k_B T$ exhibits an abrupt linear fall to large negative values with increasing temperature.

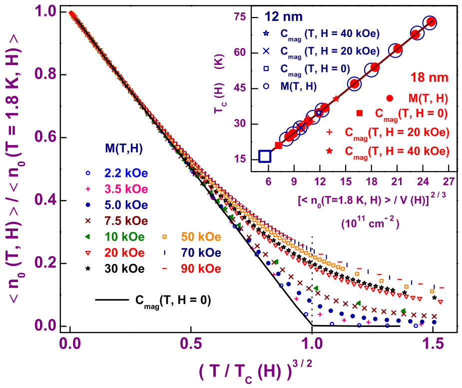

FIG. 4 (color online). Scaling of the magnon BE condensate fraction with $(T/T_c)^{3/2}$ at different fields for $d = 12$ nm. The inset highlights the scaling of $T_c(H)$ with $[\langle n_0(T = 1.8\ \mathrm{K}, H)\rangle / V(H)]^{2/3}$.

As a consistency check, the field-induced change in magnetic entropy, ΔS_{mag}, is determined from $M_H(T)$ using the Maxwell thermodynamic relation $(\partial S_{\mathrm{mag}}/\partial H)_T = (\partial M/\partial T)_H$. The top inset of Fig. 2 displays the plots of $(\Delta S_{\mathrm{mag}})_T = [S_{\mathrm{mag}}(H) - S_{\mathrm{mag}}(H = 0)]_T = \int_0^H (\partial M/\partial T)_H (dH)_T$ against $[T/T_c(H = 0)]^{3/2}$ at a few fixed fields, with $T_c(H = 0) = 16.5(5)$ K [20.5(5) K] for $d = 12$ nm [18 nm] obtained from $c_{\mathrm{mag}}(T, H = 0)$. In striking agreement with the characteristic BEC behavior of entropy, $\Delta S_{\mathrm{mag}}(T)$ decreases with temperature as $T^{3/2}$ in the limit $T \to 0$ so as to approach zero particularly when $H = 0$. Since the magnon condensate fraction (with zero entropy) reduces with increasing field (due to the destruction of phase coherence by H), the value of $\Delta S_{\mathrm{mag}}(T)$ at $T = 0$ increases with H. Furthermore, $-\Delta S_{\mathrm{mag}} = S(H = 20\ \mathrm{kOe}) - S(H = 0)$, calculated from the specific heat $C_H(T)$ data recorded at $H = 0$ and $H = 20$ kOe, conforms well with that determined from $M(T, H = 20\ \mathrm{kOe})$. Such a perfect agreement between the two sets of data is found at other fields as well.

Next, *nc*-Gd is compared with the systems that are known to undergo BEC of magnons. In spin-gap or spin-dimer compounds (yttrium iron garnet films), the applied magnetic field (microwave parametric pumping) tunes the system to the quantum critical point (critical quasiequilibrium magnon density) so as to induce BEC. By contrast, as is the case for an ideal BEC, the temperature drives the BEC transition in *nc*-Gd. The applied field needed to induce BEC in quantum antiferromagnets is often quite large, and hence for a direct comparison with the theory, the bulk properties cannot be measured in the BEC regime with the same ease as in *nc*-Gd, where a true BEC phase transition occurs in zero field (Fig. 4). This makes *nc*-Gd a unique system in which the BEC picture can be put to a stringent test. Moreover, as in *nc*-Gd, the deviations from the conventional spin-wave behavior of magnetization at low temperatures, observed previously in elongated Fe nanoparticles [25], ferrite nanoparticles [26], and Co-Pt nanopillars [27], could well be the manifestation of the BEC of magnons. Thus, nanocrystalline or nanostructured magnets may form an entirely new class of magnon BEC systems.

To summarize, our observations, such as (i) the critical temperature $T_c \sim H^{2/3}$, (ii) at $H = 0$, the condensate fraction scales with $[T/T_c]^{3/2}$ right up to $T = T_c$ such that it possesses the value zero at $T \geq T_c$, (iii) $T_c(H) \propto$ (field-dependent condensate density)$^{2/3}$, (iv) $\mu \to 0$ as $H \to 0$ for $T \leq T_c$ and abruptly falls to large negative values as the temperature exceeds T_c, and (v) the magnetic field-induced change in the magnon entropy follows the $T^{3/2}$ power law at low temperatures and goes through a peak at $T_c(H)$, amply corroborate the BEC of magnons at low temperatures in *nc*-Gd.

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[1] T. Matsubara and H. Matsuda, Prog. Theor. Phys. **16**, 569 (1956); I. Affleck, Phys. Rev. B **43**, 3215 (1991).
[2] T. Giamarchi and A. M. Tsvelik, Phys. Rev. B **59**, 11 398 (1999).
[3] C. D. Batista and G. Ortiz, Phys. Rev. Lett. **86**, 1082 (2001); Adv. Phys. **53**, 1 (2004).
[4] M. Jaime *et al.*, Phys. Rev. Lett. **93**, 087203 (2004).
[5] A. A. Aczel *et al.*, Phys. Rev. Lett. **103**, 207203 (2009).
[6] A. A. Aczel *et al.*, Phys. Rev. B **79**, 100409(R) (2009); M. Kofu *et al.*, Phys. Rev. Lett. **102**, 177204 (2009).
[7] A. Oosawa *et al.*, Phys. Rev. B **66**, 104405 (2002).
[8] T. Nikuni *et al.*, Phys. Rev. Lett. **84**, 5868 (2000).
[9] C. Rüegg *et al.*, Nature (London) **423**, 62 (2003).
[10] C. Rüegg *et al.*, Phys. Rev. Lett. **93**, 037207 (2004).
[11] B. S. Conner *et al.*, Phys. Rev. B **81**, 132401 (2010).
[12] T. Radu *et al.*, Phys. Rev. Lett. **95**, 127202 (2005).
[13] L. Yin *et al.*, Phys. Rev. Lett. **101**, 187205 (2008); V. S. Zapf *et al.*, Phys. Rev. Lett. **96**, 077204 (2006).
[14] A. I. Bugrij and V. M. Loktev, Low Temp. Phys. **33**, 37 (2007).
[15] S. O. Demokritov *et al.*, New J. Phys. **10**, 045029 (2008).
[16] V. N. Glazkov *et al.*, Phys. Rev. B **69**, 184410 (2004).
[17] S. O. Demokritov *et al.*, Nature (London) **443**, 430 (2006).
[18] D. Snoke, Nature (London) **443**, 403 (2006).
[19] A. V. Chumak *et al.*, Phys. Rev. Lett. **102**, 187205 (2009).
[20] R. Kruk *et al.*, Phys. Rev. B **73**, 054420 (2006).
[21] S. P. Mathew *et al.*, J. Phys. Conf. Ser. **200**, 072047 (2010).
[22] T. Izuyama and R. Kubo, J. Appl. Phys. **35**, 1074 (1964).
[23] J. W. Cable, R. M. Nicklow, and N. Wakabayashi, Phys. Rev. B **32**, 1710 (1985).
[24] O. Nohadani *et al.*, Phys. Rev. B **69**, 220402 (2004).
[25] P. Crespo *et al.*, Phys. Rev. B **69**, 012403 (2004).
[26] K. Mandal *et al.*, Europhys. Lett. **75**, 618 (2006).
[27] L. H. Bennett and E. Della Torre, Physica (Amsterdam) **403B**, 324 (2008).