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Low-loss barium ferrite quasi-single-crystals for microwave application

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Barium hexaferrites ($\text{BaFe}_{12}\text{O}_{19}$) are especially useful for microwave/millimeter devices. Due to large ferromagnetic resonance (FMR) loss (linewidths >2 kOe), traditional compacts of polycrystalline Ba ferrites indeed hinder the utilization of the materials for practical devices. The present experiment demonstrates that the quasi-single-crystal Ba ferrite disks can be fabricated by a single solid-state reaction technique without liquid phase participation, combining with a processing of alignment for the ferrite seed crystals. The ferrite bulks show a pure hexagonal Ba ferrite phase, an expected $4\pi M_s$ of 4.48 kG, and coercivity of $10\sim 20$ Oe along the c axis, similar to the results of a typical single crystal. The FMR measurement indicates that the sample yields an anisotropy field of 16.0 kOe and a linewidth of about 300 Oe at U -band frequencies. Although the linewidth is broader than ideal Ba ferrite single crystals ($\Delta H < 100$ Oe), it may be possible to reduce to 100 Oe by eliminating pores, cracks, local grain boundary, and nonuniformity. In terms of material preparation, we believe that it is cost effective in the production of future microwave devices. © 2007 American Institute of Physics. [DOI: [10.1063/1.2709726](https://doi.org/10.1063/1.2709726)]

I. INTRODUCTION

Barium hexaferrites [M -type $\text{BaFe}_{12}\text{O}_{19}$ ($\text{Ba}M$)] are especially useful for microwave devices, permanent magnets, and magnetic recording materials.¹ In the past several decades, a number of experimental investigations on low ferromagnetic resonance (FMR) loss single-crystal microwave ferrites, such as $\text{Ba}M$, $\text{Sr}M$ hexaferrite, spinel, and garnet yttrium aluminum garnet (YIG) ferrites, have indicated that these materials are of interest in microwave device applications, e.g., circulators, filters, isolators, inductors, and phase shifters. However, the growth of single-crystal ferrites is slow, complex, and expensive.^{2,3} On the other hand, although traditional polycrystalline ferrite compacts can easily be produced, large FMR losses (e.g., linewidths >2 kOe for $\text{Ba}M$ polycrystalline ferrite⁴) indeed hinder the utilization of the materials for practical devices.

In this paper, a detailed study of growth processing and ferromagnetic resonance measurement of quasi-single-crystal Ba ferrite is presented. Importance is that the quasi-single-crystal ferrite is obtained by solid-solid growth technique, rather than by traditional solid-liquid equilibrium method, such as Bridgeman-Stockbarger technique,^{5,6} Czochralski technique,⁷ and zone-melting technique.⁸ Although the crystal growth by solid-liquid equilibrium is probably the most widely used commercial process for single-crystal growth, high costs are considerable and inevitable. In comparison to other growth processes, the main advantages of this solid-solid growth method include the following: it permits growth at low temperatures without the presence of additional com-

ponents, the shape of the grown crystal is fixed beforehand so that some crystals having complicated shape are easily grown, and its orientation can be easily controlled. In addition, the level of impurities and other additional components can be reduced.⁹ Therefore, the technique may have potential to be used in industrial production of magnetic oxide crystals, since this growth process is relatively simple and readily controllable.

II. EXPERIMENT

In the preparation of the quasi-single-crystal, the starting material of stoichiometric $\text{BaFe}_{12}\text{O}_{19}$ particles (diameter: $0.5\sim 0.8$ μm) were provided by TransTech, Inc. To achieve high quality quasi-single-crystal blocks, the initial particles were reduced to $0.3\sim 0.5$ μm by ball milling. This was an important step in obtaining the quasi-single-crystals. These particles or seed grains were aligned by the application of an external magnetic field of 10 kOe and a pressure of 1500 psi. The magnetic field direction was parallel to the applied stress. Then, the “green” sample of disk shape ($D=10$ mm and $t=2$ mm) was dried and fired in air for a soak time of 15 h at temperatures ranging from 1300 to 1400°C. Finally, the sample was cooled down to room temperature at 50°C/h.

The samples were characterized in terms of their structural, magnetic, and microwave properties. Crystallographic orientation information was obtained from Philips X'pert PRO x-ray diffraction (XRD) measurements using $\text{Cu } K\alpha$ radiation source. The morphology of the samples was examined using a Hitachi S-4800 ultrahigh resolution scanning electron microscope (SEM). Static magnetic properties were measured using a vibrating sample magnetometer (VSM).

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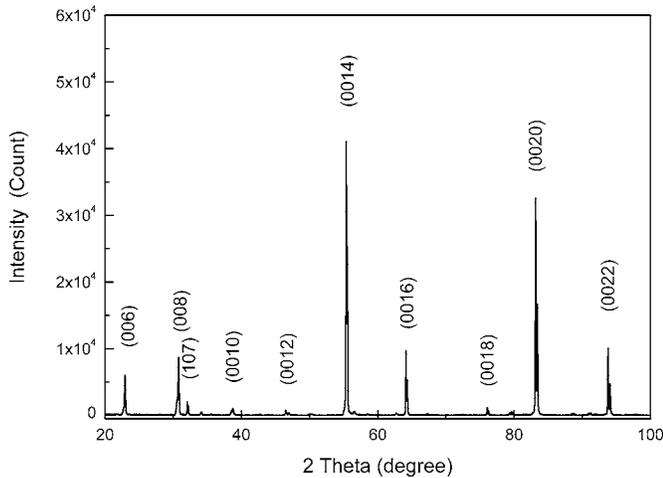


FIG. 1. X-ray diffraction pattern of a $\text{BaFe}_{12}\text{O}_{19}$ (BaM) quasi-single-crystal.

The disk was sliced to rectangular plates ($L=3$ mm, $W=2$ mm, and $t=0.05\sim 0.2$ mm) for use in FMR measurements. An external magnetic field (H_0) parallel to the c axis was applied out of the sample plane for the FMR measurements using a TE_{01} mode propagation in a U -band ($42\sim 55$ GHz) waveguide.

III. RESULTS AND DISCUSSION

Crystallographic, morphologic, and static magnetic measurements provide clear and conclusive evidences of high quality quasi-single-crystal with hexagonal structure and magnetic properties which can be in agreement with a BaM single crystal. The detected diffraction peaks almost come from the c -plane scattering except for a weak (107) face. These identified main peaks include (006), (008), (0010), (0012), (0014), (0016), (0018), (0020), and (0022), as shown in Fig. 1. The diffraction peaks, indexed to $(0,0,2n)$, are consistent with the preferential orientation of c -axis crystals perpendicular to the disk plane.

Figure 2 presents a morphology of the natural surface of a disk. It is clearly observed that many growth profiles of the hexagonal crystals spiral on the surface of the sample, which reflects a crystal growth process of high quality. A single crystal or quasi-single-crystal can be formed by continuing growth of the single-crystal region into the rest of the

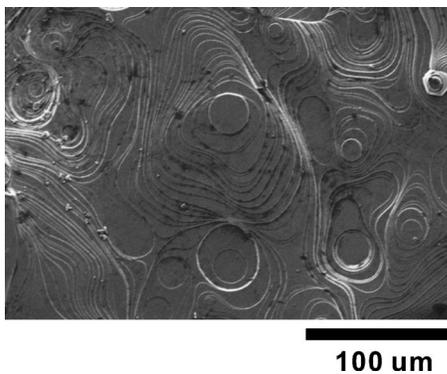


FIG. 2. SEM micrograph for the surface of a BaM ferrite quasi-single-crystal.

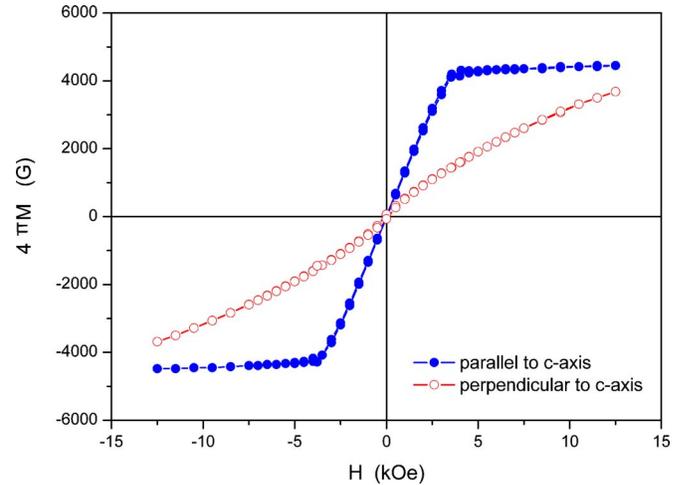


FIG. 3. Hysteresis loops of Ba ferrite quasi-single-crystal when an external field is parallel and perpendicular to the c axis.

sample, starting from those seed grains. Generally, in this monocomponent solid-solid equilibrium reaction, driving force for grain growth during sintering is mainly caused by three sources: residual strain, orientation effects, and grain-size effects. The change ΔG in the Gibbs free energy during a process in a crystal can be described below as⁹

$$\Delta G = w - q + G_s + \Delta G_0, \quad (1)$$

where w is the work done either in deliberate straining or in fabrication, q is the energy released as heat, G_s is the surface free energy of the grain, and ΔG_0 is the difference in free energy between the grain orientation existing in the material and the free energy of some other orientation. Most of w resides in the dislocation arrays associated with grain boundary. Thus, grain boundaries also contribute excess free energy due to their surface energy. Obviously, small grains have high solubility due to the higher surface energy G_s that provides important contributions to the driving force for recrystallization. Since grain-boundary energy depends on the relative orientation of the two grains, the present process reasonably has a unique ΔG_0 due to aligned grains. Some crystallites with low energy will tend to grow at the expense of those unfavorably oriented. In our case, however, orientation and grain size effects are important because large strains are not possible in oxide materials.

The magnetometry measurements indicate that the saturation magnetization, $4\pi M_s$, was measured to be 4.48 kG which corresponds to bulk values reported in the literature.¹⁰ The quasi-single-crystal of BaM exhibits a coercivity of $10\sim 20$ Oe along the c axis, whereas the remanence is nearly zero due to the large demagnetizing fields, as presented in Fig. 3. It is worth noting that the in-plane (hard axis) magnetization deviates from linearity for small fields, which is attributed to locally inhomogeneous growths in the crystals. Nevertheless, the hysteresis loops are similar to those reported in single-crystal film grown by liquid phase epitaxy (LPE).¹¹

Figure 4 shows the FMR measurements of frequency as

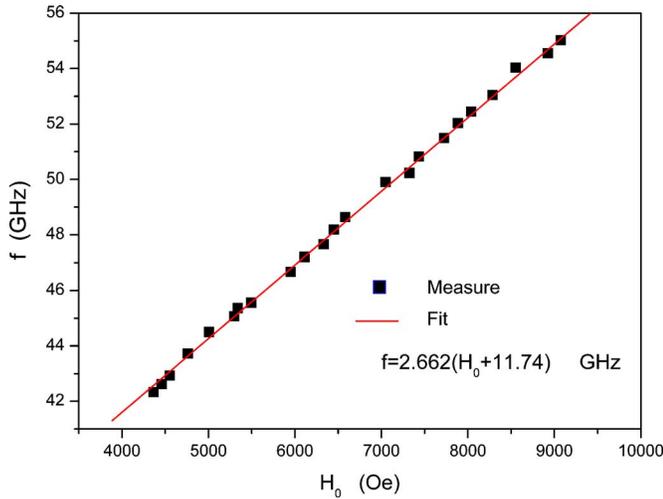


FIG. 4. Dependence of FMR frequency on an external field (H_0) for a BaM quasi-single-crystal with a thickness of $100\ \mu\text{m}$.

a function of external field (H_0) for the sample. Since H_0 is normal to the sample, the FMR condition is given as follows:¹²

$$f = \gamma' (H_0 + H_A - N_z M), \quad (2)$$

where f is the resonance frequency in gigahertz, H_0 is the resonance external field in kilo-oevsted, and $\gamma' = 2.662\ \text{GHz/kOe}$ (the Lande factor $g = 1.90 \pm 0.02$). Here, an anisotropy field can be deduced to be $H_A = 16.01\ \text{kOe}$, assuming demagnetizing factor $N_z = 0.95$. These results are reasonable for a BaM crystal.

The FMR linewidth ΔH is plotted as a function of frequency from 42 to 55 GHz, as illustrated in Fig. 5. The FMR linewidths of the measured samples (thickness of $\sim 100\ \mu\text{m}$) scale from 320 to 680 Oe at the U -band frequency. The low-

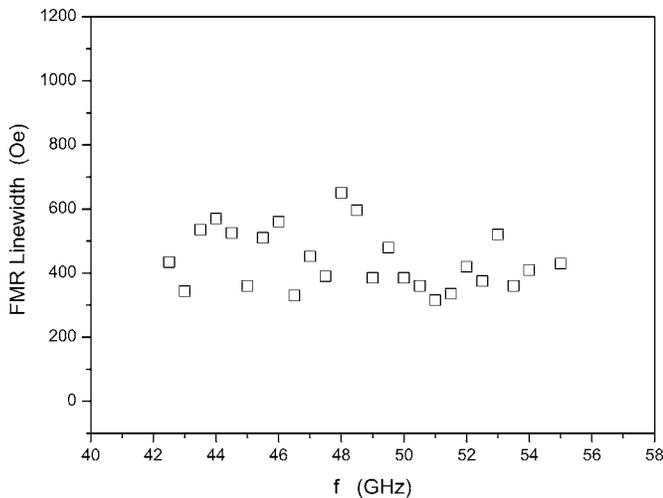


FIG. 5. FMR linewidth vs frequency at the frequency range of 42–55 GHz.

est ΔH of the BaM quasi-single-crystal is 320 Oe at 53 GHz, much lower than those ($\Delta H > 2000\ \text{Oe}$) for BaM polycrystalline compacts.⁴ However, an ideal single crystal¹³ and LPE thick films ($23\ \mu\text{m}$ thick)¹¹ exhibited very low FMR linewidths of 30–40 Oe. Clearly, although the linewidths for these quasi-single-crystals are still more than that of ideal Ba ferrite single crystals ($\Delta H < 100\ \text{Oe}$), the results ($\Delta H \sim 300\ \text{Oe}$) indicate that the materials are suitable for applications of practical devices at U -band frequencies. From our optical observation, this material contains some pores, cracks, local grain boundary, randomly oriented crystallites, and nonuniformity, which explains the broadening of the linewidths.^{14,15} Therefore, we believe that the linewidth can be lowered down to 100 Oe by modifying starting particle size, improving magnetic alignment of the powders under a pressure, and finally optimizing sintering process.

IV. CONCLUSION

The work presents a solid-solid equilibrium sintering technique used to prepare quasi-single-crystal barium hexaferrite with low microwave loss. We have developed a processing scheme in which Ba hexaferrite is produced having good crystallography, surface morphology, static magnetic properties similar to those of a BaM single crystal, and low FMR linewidths ($\sim 300\ \text{Oe}$). These properties make these quasi-single-crystal Ba ferrites ideal for use at U -band frequencies, instead of expensive single crystal. Therefore, the utilization of the materials has potential to reduce costs for microwave devices.

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