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S. G. Wang

S. D. Yoon

C. Vittoria
Northeastern University

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Microwave and magnetic properties of double-sided hexaferrite films on (111) magnesium oxide substrates

S. G. Wang, S. D. Yoon, and C. Vittoria^{a)}

Department of Electrical and Computer Engineering, Northeastern University, Boston, Massachusetts 02115

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We have deposited hexaferrite films of $\text{BaFe}_{12}\text{O}_{19}$ on both sides of a magnesium oxide, (111) MgO, substrate by the liquid phase epitaxy (LPE) deposition technique. The procedure entailed the deposition of seed layers of $\text{BaFe}_{12}\text{O}_{19}$ onto the substrate by laser ablation deposition and then dipping of the substrate into a molten flux. Whereas the seed layers were the order of $0.5\ \mu\text{m}$, the total thickness after LPE deposition was $\sim 45\ \mu\text{m}$. The ferrimagnetic resonance linewidth at 60 GHz, 27 Oe, is as low as it has ever been measured before in $\text{BaFe}_{12}\text{O}_{19}$ to the best of our knowledge. The static field measurements of the saturation magnetization and anisotropy field were in agreement with previous measured bulk values. © 2002 American Institute of Physics.

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I. INTRODUCTION

The deposition of magnetic films on two sides of a substrate was initiated about 20 years ago. The purpose of double-sided growth deposition was to grow very thick films of yttrium-iron-garnet (YIG) on gadolinium-gallium-garnet (GGG) substrates. For example, thickness on the order of $100\ \mu\text{m}$ (total of $200\ \mu\text{m}$) was possible by depositing YIG films on GGG substrates by the liquid phase epitaxy (LPE) deposition technique. The rationale for depositing double-sided films was that the strain induced during growth on the two surfaces of the substrates would cancel each other sufficiently to allow thicker film deposition of YIG. The growth of single layers of YIG on GGG induced strain² between the film and substrate due to the thermal expansion coefficient differential between the film and substrate. Typically, films of the order of $10\text{--}15\ \mu\text{m}$ would peel off or craze or simply crack under the strain. The strain induced by the lattice constant mismatch between a YIG film and GGG substrate was negligible compared to the strain induced from the thermal expansion differential. Surface quality, defects, grooves on the surface, etc. conceivably can also induce strain. This source of strain can be dismissed if the surface is reasonably polished. By depositing YIG on two surfaces of GGG it was then possible to induce two strain fields which approximately canceled each other.

We have extended the above idea¹ to the growth of hexaferrite films of $\text{BaFe}_{12}\text{O}_{19}$ on both surfaces of a (111) magnesium oxide (MgO) substrate. Glass *et al.*³ were able to deposit single layers of $\text{BaFe}_{12}\text{O}_{19}$ as thick as $10\text{--}16\ \mu\text{m}$ on (0001) sapphire (Al_2O_3) substrates. A buffer layer or seed layer of $\text{BaFe}_{12}\text{O}_{19}$ was deposited between the film and substrate. The seed layer was deposited by the ion beam sputtering technique. Deposition of $\text{BaFe}_{12}\text{O}_{19}$, $\text{SrFe}_{12}\text{O}_{19}$, and $\text{PbFe}_{12}\text{O}_{19}$ single layers on substrates of $\text{SrGa}_{12}\text{O}_{19}$ was accomplished⁴ by Dötsch *et al.* In their work a buffer layer was not needed, since the lattice constants and thermal ex-

pansion coefficients were reasonably matched between the film and substrate. Nevertheless, thickness of $10\text{--}15\ \mu\text{m}$ was achieved³ utilizing the LPE technique. Again, the strain induced during deposition was too great to increase the thickness beyond $15\ \mu\text{m}$. We have extended the work of the above researchers¹⁻⁴ by depositing films of $\text{BaFe}_{12}\text{O}_{19}$ with thickness greater than $15\ \mu\text{m}$. We have adopted the lessons learned from the YIG work of double-sided deposition^{1,2} and modified the technique developed by Glass *et al.*³ for the growth of single layers of $\text{BaFe}_{12}\text{O}_{19}$. Specifically, we report the deposition of double-sided layers of $\text{BaFe}_{12}\text{O}_{19}$ on substrates of (111) MgO. Seed layers of $\text{BaFe}_{12}\text{O}_{19}$ ($0.5\ \mu\text{m}$) were deposited on both sides of the substrate using the laser ablation deposition technique. This is a departure from Glass *et al.*'s³ procedure, since buffer layers as deposited require no postannealing. Also, we used substrates of MgO instead of sapphire. The lattice constant match between the film and substrate improved, but not the thermal expansion coefficients of the two materials. The point of this work was to minimize strain effects due to thermal expansion differentials between the substrate and film by depositing two-sided films on the MgO substrate. In Sec. II we describe the preparation of the seed or buffer layer, LPE technique, and material preparation as related to the overall growth procedure. In Sec. III, x-ray, scanning electron microscope (SEM), vibrating sample magnetometer (VSM), and ferrimagnetic resonance (FMR) characterization measurements are presented. In Sec. IV, there is discussion and our conclusions are summarized.

II. MATERIAL PREPARATION

The deposition of double-sided films of $\text{BaFe}_{12}\text{O}_{19}$ on a substrate of (111) MgO involved the following step-by-step procedure.

A. Deposition of seed layers

Substrates of (111) MgO were utilized, since the lattice constant mismatch between the film and substrate was mini-

^{a)}Electronic mail: vittoria@research.neu.edu

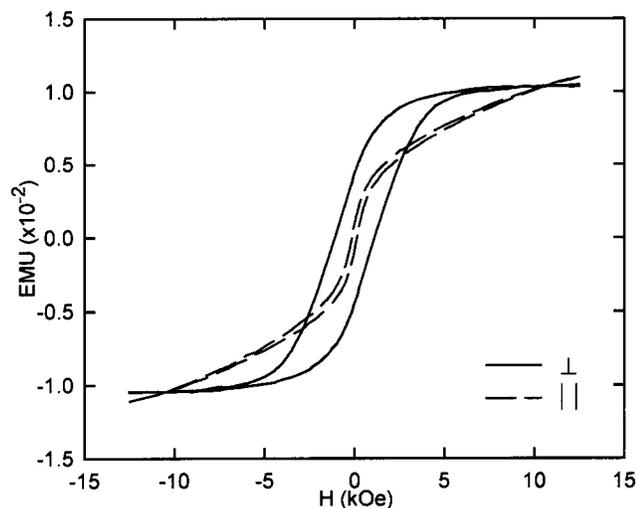


FIG. 1. VSM hysteresis loop behavior of a double-sided $\text{BaFe}_{12}\text{O}_{19}$ seed layer on (111) MgO deposited by LAD. The solid line corresponds to an out-of-plane VSM measurement and the dashed line to an in-plane VSM measurement.

mal. The lattice constant of $\text{BaFe}_{12}\text{O}_{19}$ in the basal plane was 5.89 vs 5.95 Å for (111) MgO. This implies that growth was induced with the c axis normal to the film or substrate plane. Both surfaces of MgO were polished, since deposition was required on both surfaces. In normal deposition of a single layer of $\text{BaFe}_{12}\text{O}_{19}$ by the laser ablation deposition (LAD) technique one would coat the backside of the substrate with silver paint in order to insure thermal contact with the heater. We avoided this step by simply placing the substrate in physical contact with the heater. Removal of silver paint is essential in preparing a surface that is free of impurities and defects. The reader can refer to Ref. 5 for a description of the LAD technique for deposition of single layers of ferrites. After depositing one seed layer of 0.5 μm on one side of MgO, we flipped the side over and deposited another seed layer on MgO. In Fig. 1 the VSM measurement of a typical seed layer of 0.5 μm thick is shown. The symbols \perp denote that the external field is applied perpendicular to the film plane and \parallel parallel to the film plane. The fact that the hysteresis loops exhibited curvature at saturation fields for the \perp case and the coercive field was 1.0 kOe implied a nonuniform distribution of the demagnetizing field and defects in the film, indicating poor film quality. Nevertheless, the c axis was on average aligned perpendicular to the film plane. X-ray and VSM measurements confirmed the c -axis alignment.

B. Film growth by the LPE technique

We used barium-based flux for LPE growth, since it is much less volatile than bismuth- or lead-based flux, and thus avoided contamination by barium ferrite from those cations. The $\text{BaFe}_{12}\text{O}_{19}$ flux melt was prepared from a mixture of iron oxide (Fe_2O_3), barium carbonate (BaCO_3), and boron oxide (B_2O_3). The ratio of the powder weights of Fe_2O_3 , BaCO_3 , and B_2O_3 relative to each other was chosen to be $x:1:0.571$, where x ranged from 0.24 to 0.26. The powders were ground and mixed homogeneously. Based on the

chemical formula of $\text{BaFe}_{12}\text{O}_{19}$, the ratio between the powder weights of Fe_2O_3 and BaCO_3 was fixed. The melt consisted of a mixture of barium oxide (BaO) and B_2O_3 ,³ since the carbonates evaporated.

A 150 mL platinum crucible was used to hold the melt. The starting amount of powder mixture occupied two thirds of the volume of the crucible. The crucible was replenished with about one fourth of the initial weight mixture as the carbonates decomposed into CO_2 gas and evaporated. The crucible was heated to 1200 °C in an enclosed furnace. After decomposition of the carbonates was complete, the crucible was replenished with small amounts, of the order of 10–20 g, of the powder mixture as it was heating. Pellets of the powder mixture were added only after the bubbling ceased. The bubbling was caused by the evaporation of CO_2 gas. The melting procedure was closely monitored in order to avoid overflow of the melt. After all of the powder mixture was melted, it was heated again to 1150 °C for 8–12 h, to ensure a uniform melt. At this point we assumed that the hexaferrite composition was complete and that it was in a liquid state in the molten flux. Finally, the melt was cooled down to room temperature.

Prior to dipping the substrate into the melt, the melt was stirred with a substrate holder made of platinum wires at 1100 °C. After 2 h the holder was lifted out of the furnace, and the temperature of the furnace was reduced slowly to between 850 and 920 °C. The growth temperature ranged between 850 and 920 °C. Generally speaking, the higher the content of iron, the higher the saturation temperature, or the cooling down temperature. For growth temperatures below 830 °C, for example, nonmagnetic crystals (probably barium borate) were predominant in the growth of the film on the substrate.

When the temperature had been cooled to the desired growth temperature, the substrate was lowered into the melt for deposition. The substrate was preheated at a position about 5–10 mm above the melt for about 10 min. The temperature of the melt was maintained at a constant value or at a slowly decreasing temperature during growth. The typical growth temperature was 850–920 °C. It should be noted that spontaneous crystallization on the holder should be avoided. A large amount of spontaneous crystallization greatly reduced the growth rate, and also degraded the quality of the films. The growth rate was typically about 5–6 μm per hour. The substrate was rotated at about 30 rpm during growth in order to avoid depletion of the hexaferrite melt content in the crucible, and to ensure homogeneous temperature of the melt. After removal of the substrate from the melt, the substrate was cleaned in a solution of dilute nitric and acetic acid to remove nonferrite residues. The ferrite films were then characterized by x-ray diffraction (XRD), SEM, VSM, and FMR measurements. For the VSM and FMR measurements we mechanically polished away one layer so that we could examine one layer at a time. Interestingly, the surface of the substrate exposed to the bottom of the crucible grew a thicker layer than did the other surface of the substrate.

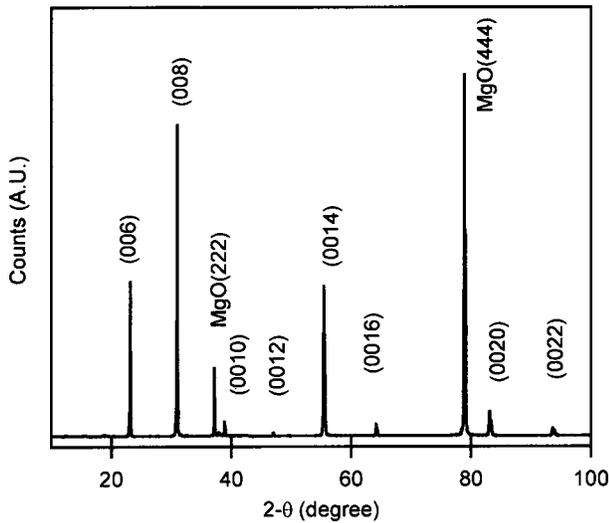


FIG. 2. X-ray diffraction pattern of a 25 μm thick $\text{BaFe}_{12}\text{O}_{19}$ film on (111) MgO grown by LPE.

III. CHARACTERIZATION MEASUREMENTS

A. X-ray measurements

The film grew with the c axis normal to the substrate plane [(111) plane]. This finding was confirmed from x-ray diffraction measurements using $\text{Cu } K\alpha$ radiation and a conventional diffractometer equipped with a goniometer. Measurements of multiple orders of diffraction from atomic planes parallel to the substrate surface were used to verify the hexagonal structure, shown in Fig. 2. SEM was used to examine the surface of a typical layer, shown in Fig. 3. An

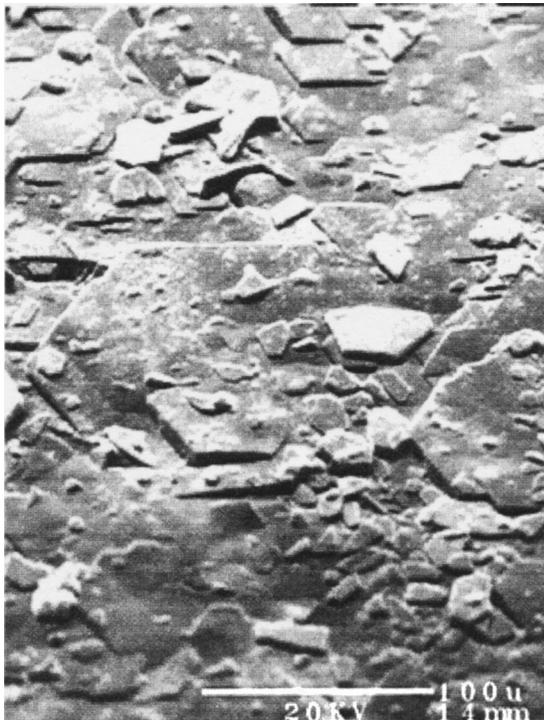


FIG. 3. SEM image of a 25 μm thick $\text{BaFe}_{12}\text{O}_{19}$ film on (111) MgO grown by LPE.

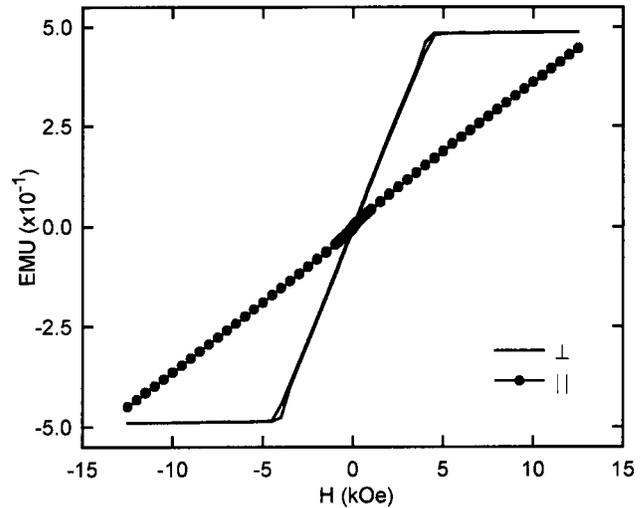


FIG. 4. VSM hysteresis loop behavior of a double-sided $\text{BaFe}_{12}\text{O}_{19}$ film on (111) MgO grown by LPE. The solid line corresponds to an out-of-plane VSM measurement and the dashed line to an in-plane VSM measurement.

hexagonal platelet can be seen on the surface of the substrate, further confirming that the surface was indeed the basal plane of the hexagonal structure.

B. Magnetic characterization measurements

The magnetic moment measured as a function of the external field H by the VSM is shown in Fig. 4. H was applied perpendicular to and in the plane of the film. In this measurement the total thickness of the layer was 23 μm . The coercive field was less than 10 Oe. As one would expect from a single-crystal hexaferrite, the remanence was nearly zero due to the large demagnetizing fields. It can be noticed that for H in the film plane there is a small barely perceptible curvature of the M vs H curve near $H=0$. The curvature is a result of there being a small amount of ferrite deposited along the edge of the substrate, since bulk single crystals have lower H_c . For example, VSM data of bulk single crystal platelets did not exhibit such a curvature. The saturation magnetization, $4\pi M_s$, was measured to be 4.4 kG which corresponds to bulk values reported in the literature.⁶ The uniaxial magnetic anisotropy field, H_A , could not be measured with our VSM equipment, since the maximum field available to us was 13 kOe which was less than H_A of $\text{BaFe}_{12}\text{O}_{19}$.⁶

However, we were able to measure H_A utilizing FMR. In Fig. 5 we have plotted the FMR frequency as a function of H for H applied perpendicular to the film plane or parallel to the c axis. FMR was performed on one of the layers (~ 23 μm thick) deposited on (111) MgO . The other layer deposited on the other surface was mechanically removed. For fields above saturation ($4\pi M_s \cong 4.4$ kOe), the measured resonance dispersion obeys the following relation:⁷

$$f = \gamma'(H + H_A - NM_s),$$

where f is the resonance frequency in GHz, and H is the resonance external field in kOe, and $\gamma' = 2.788$ GHz/kOe. If we identify $H_A - 4\pi M_s = 11.938$ kOe, where $4\pi M_s$

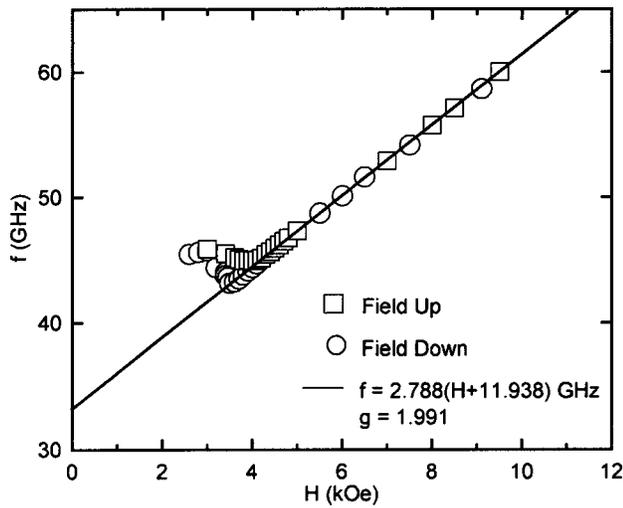


FIG. 5. FMR frequency vs external magnetic field.

=4.4 kG from the VSM measurement, $f = 2.788 (H + 11.938)$. Thus, $H_A \approx 16.4$ kOe. We deduced the g factor as $g = 1.991 \pm 0.002$.

For fields below saturation, magnetic remanence depends on the approach to saturation, giving rise to microwave resonance hysteresis. Resonance data observed for increasing H fields during FMR experiments are marked by circles. For H swept in a decreasing manner (from high to low fields) data are shown by squares in the plot. The microwave hysteresis behavior observed is the result of nonlinear⁷ resonance excitations in the multidomain configurations for H below saturation fields of 4.4 kOe.

Finally, the FMR linewidth, ΔH , is plotted as a function of frequency from 43 to 60 GHz, as shown in Fig. 6. We see that the average ΔH is about 30 Oe. The lowest ΔH of the LPE film was measured to be 27 Oe at 56 GHz, seen in Fig. 7. The linewidth of single crystal spheres of $\text{BaFe}_{12}\text{O}_{19}$ was measured⁸ to be ~ 35 Oe at 75 GHz. It is interesting to note that ΔH was⁹ also measured to be independent of the fre-

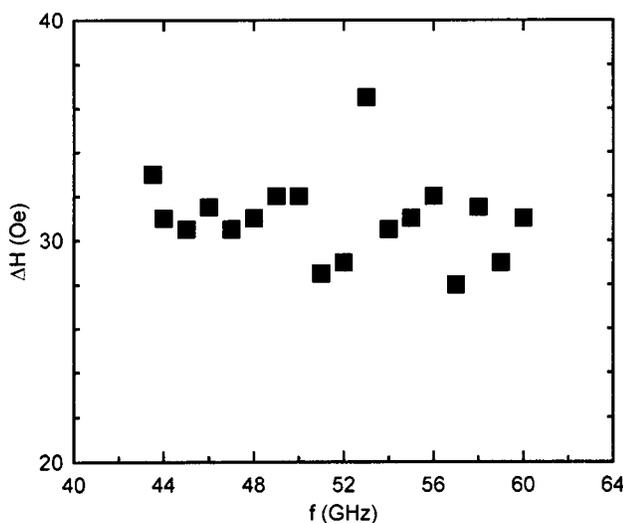


FIG. 6. FMR linewidths vs frequency.

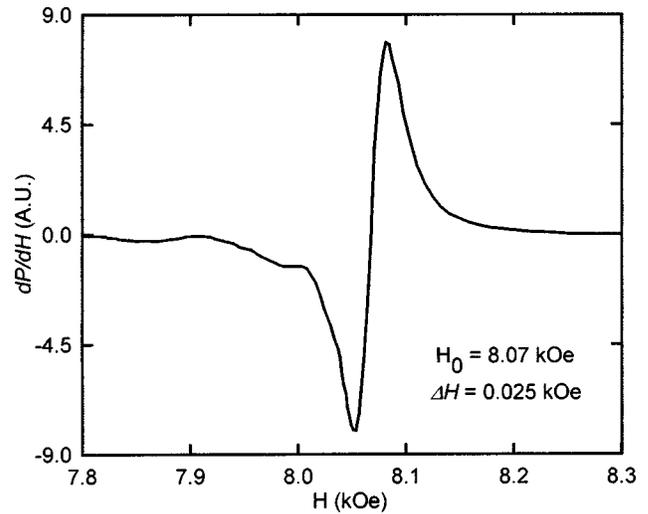


FIG. 7. FMR frequency spectrum at 56 GHz.

quency from 50 to 110 GHz. FMR linewidth measurements by Karim *et al.*⁸ indicated that ΔH increased from 25 to 45 Oe as the frequency was varied from 60 to 90 GHz. However, with a small amount of Mn impurity ($\sim 0.8\%$) added to the chemical composition of barium ferrite it altered the slope of ΔH versus frequency significantly. It appears that starting materials and the purity of the melt may be very critical in the magnetic relaxation process of hexaferrite materials, especially in M -type hexaferrites.

IV. DISCUSSION AND CONCLUSION

It appears that it may be possible to deposit thick double layers of $\text{BaFe}_{12}\text{O}_{19}$ or M -type hexaferrites on (111) substrates of MgO. A total thickness of 45–50 μm of sufficiently good quality was achieved. For practical device fabrication we would require thickness of the order of 100 μm . However, thicker films imply longer growth time. For example, 100 μm thick films (50 μm on both sides of the substrate) imply growth time of the order of 8–10 h. For industrial use the growth condition should be improved in order to reduce the growth time. In addition to MgO substrates we used sapphire, quartz, and alumina substrates. We did not succeed in growing $\text{BaFe}_{12}\text{O}_{19}$ films on these substrates by the LPE/LAD technique. However, the use of a sapphire substrate presents somewhat of a paradox. Glass *et al.*³ succeeded in growing, by the LPE technique, a single layer of 16 μm thick $\text{BaFe}_{12}\text{O}_{19}$ on a seed layer deposited on (0001) sapphire substrates. The seed layer of $\text{BaFe}_{12}\text{O}_{19}$ was deposited by ion beam sputtering and its thickness was about 0.5 μm . We are left with the conclusion that growth of $\text{BaFe}_{12}\text{O}_{19}$ layers on (0001) sapphire is critically dependent on the quality or growth details of the seed or buffer layer. Furthermore, since ion beam sputtering implies oxygen deficiency in the seed layer, it implies that LPE growth of $\text{BaFe}_{12}\text{O}_{19}$ films on (0001) sapphire is enhanced by the use of seed layers with oxygen deficiency! These data should be compared with data for our recent success in growing thick layer of $\text{BaFe}_{12}\text{O}_{19}$ ($\sim 70 \mu\text{m}$) on (10 $\bar{1}$ 0) sapphire substrates in which the c axis was in the film plane.¹⁰ It is not clear to us why this should

be so. However, one cannot ignore the results of Glass *et al.*³ Future research should focus on the importance of the preparation of seed layers relative to the growth plane in the deposition of thicker films.

ACKNOWLEDGMENTS

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