Monodomain Theory: Experimental Verification

Abstract. The Néel thermal-activation theory of remanence in monodomain grains has been verified quantitatively in experiments on four ferrite micropowders and two natural rocks. Magnetization and demagnetization curves of thermal, isothermal, viscous, and anhysteretic remanences can all be predicted with reasonable accuracy when the Néel theory is generalized to include effects of grain interaction. Results with the natural materials indicate that interacting, single-domain grains or regions are the carriers of the magnetically hard natural remanence of some paleomagnetic rocks.

To evaluate the role of single-domain grains or regions in rocks, I have performed many experiments with thermoremanence (TRM), anhysteretic remanence (ARM), isothermal remanence (IRM), and trainage on two sets of samples: those in one set contained synthetic monodomain material; the others were fine-grained natural rocks. The observed results were compared with predictions based on the Néel (1) theory of single-domain grains. Thus I could test comprehensively the predictions of Néel theory and also determine whether the stable remanence of some rocks can be explained by monodomain theory.

The four synthetic samples contained micropowders of magnetite (Fe_3O_4) , cobalt-doped maghemite $(\gamma-Fe_{1.95}Co_{0.05}-O_3)$, hematite $(\alpha-Fe_2O_3)$, and cobalt ferrite $(CoFe_2O_4)$, dilutely dispersed in kaolin. The grain sizes of all powders were in the single-domain range.

Two rocks of paleomagnetic significance were studied: (i) a basalt thought to contain small, possibly monodomain, regions as a result of extensive alteration of magnetite grains (2); and (ii) a laterite, representative of some baked earths (3), containing extremely finegrained maghemite. The latter sample is of special interest because of its similarity to the sample used by Everitt (4) in the only previous systematic test of Néel theory.

The reason for this paucity of work is not lack of interest in the Néel theory; it is that the two basic Néel equations, which specify the conditions under which a grain or ensemble of grains is blocked (magnetically stable) and the remanence of the ensemble, involve grain volume and coercivity, and the grains in real rocks have a wide and unknown spectrum of both these parameters. I overcame this difficulty by determining the grain size-coercivity distribution of each sample (5) by a reported method (6). Use of the grain distribution as a weighting function, in application of the Néel theory to a particular sample, made computation of theoretical curves straightforward.

All experiments were performed with a ballistic magnetometer, an air-cored a-c and d-c solenoid, and a furnace combined in one instrument (7). Hysteresis properties, measured at seven to ten temperatures between 20° C and the Curie point, indicated a high proportion of single-domain material in all samples (8). The main body of experiments surveyed the distinctive thermal, isothermal, anhysteretic, and viscous remanent properties of singledomain grains (9). Here I report only the results that provide a clear-cut test of Néel theory.

Partial TRM is produced by fieldcooling over a limited temperature range; partial ARM is produced by a d-c field applied over a limited range of a-c field. Néel's interpretation of TRM and ARM as blocking processes leads to the conclusion that the sum of partial TRM's or ARM's, produced in adjoining temperature or a-c field intervals, should equal the total TRM or ARM produced over the entire interval, at least for magnetizing fields of 1 oersted or less. For larger fields, theory predicts that the contribution by high-temperature IRM, to the higher-temperature partial TRM, causes deviations from the TRM additivity law. Experimentally, the ARM additivity law was obeyed within 3 percent



Fig. 1. Isothermal remanence produced by a field of 23 oersteds at various temperatures. Sample: natural γ -Fe₂O₈.

for all samples (for $H_{d-e} = 10$ oersteds), while the partial TRM sum equalled the total TRM plus the high-temperature IRM, as predicted by theory (fields of 45 and 90 oersteds were used).

Because of the important practical implications of nonadditivity of partial TRM's [for example, in the Thellier method of paleointensity determination (10)], it is of interest to examine the spectrum of high-temperature IRM's the magnitudes of small-field IRM's produced at various temperatures. A typical experimental spectrum is shown (Fig. 1) for the sample of natural maghemite; agreement between theoretical and experimental spectra was generally good.

One of the most distinctive properties of any type of remanence is its magnetization curve, or dependence on d-c magnetizing field. Experimental ARM and TRM magnetization curves for the sample of natural magnetite (Fig. 2) rise linearly at small fields and saturate within a few hundred oersteds. Néel theory predicts much too steep an initial slope for both curves; indeed the ARM should saturate for an infinitesimal d-c field!

The experimental IRM magnetization curve rises very slowly at small fields and may not saturate except in a field of several thousand oersteds. By differentiation of the magnetization curve, the spectrum of d-c coercivities in the IRM can be obtained; agreement of such a spectrum for the sample of natural maghemite (curve c of Fig. 3) with the spectrum derived by Néel theory (curve b) is not good. Curve a shows the a-c coercivity spectrum of IRM for the same sample, obtained by differentiating the experimental a-c demagnetization curve of the saturation IRM. The theoretical spectrum, again given by curve b, is significantly different from the experimental spectrum. It appears that the coercivity of a particle in a-c fields is lower, and that in d-c fields higher, on the average, than the coercivity predicted by Néel theory.

Other experiments measured the a-c coercivity spectra of TRM's, ARM's, and IRM's produced in small, moderate, and large d-c fields; IRM's become harder—the peak in the coercivity spectrum moves to higher fields—as the magnetizing field increases, but the trend for TRM's and ARM's is just the opposite. Néel theory, however, predicts that TRM and ARM spectra should be almost independent of H_{d-e^*}



Fig. 2. Anhysteretic and thermoremanent magnetization curves. Sample: natural Fe_3O_4 .

The experimental spectra of TRM's and ARM's, produced in fields of 1 to 3 oersteds, agree almost perfectly with the spectra predicted by Néel theory, so that the deviation must be associated with moderate and large fields.

I found also that the a-c demagnetization curve of a TRM tends to be slightly softer than that of an ARM produced in the same field. The theoretically unexplained differences are not large, but seem to be significant, for they appeared for all samples examined.

Traînage decay curves (the decrease with time of a small-field IRM after the inducing field is switched off) were obtained for all samples, with the corresponding growth curves (the increase in IRM after a field is applied) for the two most viscous samples, the natural and synthetic maghemites. All traînage curves were approximately linear when plotted on a logarithmic time scale, and were in fairly close agreement with theoretical curves. The fact that the growth rate was only a few percent larger than the decay rate in the two maghemites confirmed the preponderance of single-domain material in these samples (11).

Thus Néel monodomain theory accounts quantitatively for the additivity of partial ARM's and nonadditivity of partial TRM's, for the spectrum of high-temperature IRM's, for the coercivity spectra of small-field TRM's and ARM's, for small-field traînage results, and for some relations between coercivity and blocking temperature (9). There is no consistent difference between the fits obtained with natural and with synthetic samples, the implication being that all samples contain dominantly single-domain material.

But Néel theory fails to account for

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either the magnetization curves, or the relative hardness trends in a-c demagnetization curves, of TRM's, ARM's, and IRM's. The finite initial slope of the ARM magnetization curve can be explained qualitatively (12) if one takes into account magnetostatic particle interaction, which is ignored in the Néel theory. Perhaps the other discordant results might also be explained by interaction effects.

One of the most fruitful approaches to analysis of particle interaction is that of the Preisach diagram (13), or density distribution of a and b, the coercivities of a grain in increasing and decreasing fields, respectively. In simple Néel theory, a and b are of equal magnitude, H_e , but Néel himself later showed (14) that a spatially fluctuating interaction field, H_i , could under some conditions displace the hysteresis loop of a monodomain particle an amount H_i , making $a = H_e - H_i$ and $b = -H_e$ $- H_i$.

After a-c demagnetization, an ensemble of such interacting particles is not statistically demagnetized, but is said to be polarized (15) because there is a high probability of a particular particle moment being left in the orientation or polarization favored by the local H_i vector. A particle contributes to the ensemble's remanence, or is effectively magnetized, only if it is blocked in an orientation different from its polarization. Since ARM is acquired in a d-c field which is the sum of external and interaction fields, a grain contributes to the net ARM of the ensemble only when the applied field H_{d-c} is of greater magnitude than (and opposed to) H_i . An effectively magnetized grain will revert to its polarized state, or be "demagnetized," as soon as the a-c field reaches a value equal to the numerically smaller of the switching fields a and b. Because larger values of $|H_i|$ cause |a|and |b| to become increasingly different, the softest grains in an ensemble are those with the largest values of $|H_i|$.

Many of my discordant results can be explained qualitatively by this simple model. The coercivity of a particle in a d-c field may be either *a* or *b*, and is therefore larger on the average than the a-c coercivity, which is always the numerically smaller of *a* and *b*. It is also obvious that ARM cannot saturate in infinitesimal fields, because only grains having $|H_i| < |H_{d-c}|$ contribute to an ARM, and the interactions are not normally negligible. An ARM pro-



Fig. 3. Coercivity spectra of saturation IRM. Curves a and c are experimental a-c and d-c spectra; curve b is predicted by Néel theory (a-c or d-c). Sample: natural γ -Fe₂O₃.

duced by a very small field effectively magnetizes only grains with very small H_i ; thus it has a coercivity spectrum nearly the same as that predicted by Néel theory. Larger-field ARM's will, of course, become progressively softer, since they magnetize grains with progressively larger H_i .

The thermal blocking process is analogous to the anhysteretic process except that blocking occurs at high temperatures where magnetostatic interactions are smaller because of the thermal decrease in spontaneous magnetization. Grains whose room-temperature value of $|H_i|$ is larger than $|H_{d-e}|$, but whose value of $|H_i|$ at the blocking temperature is less than $|H_{d-c}|$ will contribute to a TRM but not to an ARM. Hence the TRM induced by H_{d-e} is larger than the ARM. On the other hand, since the additional grains contributing to TRM comprise a higher $|H_i|$ fraction, TRM will be somewhat softer than ARM produced by the same field.

For quantitative testing of these ideas, experimental Preisach diagrams, or density distributions in the a,b plane, were determined experimentally by the method of Bate (16) for all samples at 20°C, for the synthetic magnetite at 470°C, and for the cobalt ferrite at 353°C. Since all grains whose representative points lie on the line $a - b = 2H_c$ on the Preisach diagram belong to an ensemble characterized by coercivity H_c when $H_i = 0$, a profile of the Preisach density distribution along such a line provides an experimental estimate of the distribution function of H_i and will be called an

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interaction profile. The fact that the half-widths of the experimental interaction profiles at 20°C were 70 to 360 oersteds showed that interactions are important in all samples.

Theoretical ARM magnetization curves were obtained by numerical integration of the experimental H_i distributions and normalization to the saturation remanence. A good match to the observations resulted (Fig. 2). Magnetization and a-c demagnetization curves of saturation IRM were well predicted from the Preisach diagram, as shown by the theoretical and experimental coercivity spectra of Fig. 3.

The TRM magnetization curve was recalculated; again I used the equations of simple Néel theory, but now took into account the fact that the total field acting on a grain, at its blocking temperature, $T_{\rm B}$, is $H_{\rm d-e} + H_{\rm i}(T_{\rm B})$. The spectrum of H_i at T_B was taken as the 20°C interaction profile suitably contracted by the thermal decrease of spontaneous magnetization between 20°C and $T_{\rm B}$. Improvement in agreement between theoretical and experimental TRM curves was spectacular (Fig. 2).

The TRM calculation is important for three reasons. It demonstrates that the properties of the monodomain grains in these samples are markedly modified by magnetostatic grain interactions. Thus it follows that two published estimates of grain size (1, 4), based on the untenable assumption that the shape of the TRM magnetization curve follows directly from Néel theory, are unreliable. Most important is the fact that the interaction spectrum, derived from the Preisach diagram, can be successfully used in calculations depending on the Néel equations; this provides a general method of analyzing the thermal properties of interacting monodomain grains.

Thus, the results of many experiments, insensitive to the interaction state of a sample, are quantitatively well predicted by the Néel theory of independent monodomain grains. Interactions are significant in some real rocks, however, and produce deviations from simple Néel theory in experiments involving the magnetization and demagnetization of remanences. Quantitative analysis in the presence of interactions can be performed by Preisach theory (isothermal processes only) or more generally by an extended form of Néel theory that takes account of interactions.

The properties of my two natural rocks agreed as well with monodomain theory as did those of the four synthetic "rocks" containing grains known to be of single-domain size. This fact strongly suggests that the natural remanence of paleomagnetic materials similar to those studied here is carried largely by high-stability, single-domain grains or regions. While problems such as development of a physically adequate model of monodomain-like regions in large grains are of great fundamental interest, it is equally important to gain further insight into the properties of monodomain assemblies and to discover what classes of rocks display such properties.

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References and Notes

- 1. L. Néel, Ann. Geophys. 5, 99 (1949). 2. E. Larson, Mituko Ozima, Minoru Ozima, T.

- L. Larson, Mittako Ozinia, J., in press.
 Nagata, D. Strangway, *Geophys. J.*, in press.
 R. L. Wilson, *ibid.* 5, 45 (1961).
 C. W. F. Everitt, *Phil. Mag.* 6, 713 (1961).
 These grain distributions are of considerable intrinsic interest and will be described elsewhere
- 6. D. J. Dunlop, J. Geomagn. Geoelec. 17, 459
- (1965) 7. G. F. West and D. J. Dunlop, in preparation.
- B. J. Dunlop, in preparation.
 —, thesis, University of Toronto (1968).
- 10. E. Thellier and O. Thellier, Ann. Geophys. 15, 285 (1959).
- 11. E. Le Borgne, ibid. 16, 445 (1960).
- E. Le Borgne, *Iola*. **10**, 445 (1960).
 D. F. Eldridge, J. Appk Phys. **32**, 247S (1961); E. D. Daniel and E. P. Wohlfarth, J. Phys. Soc. Japan **17**(B1), 670 (1962).
 F. Preisach, Z. Phys. **94**, 277 (1935).
 L. Néel, Appl. Sci. Res. **B4**, 13 (1954).
 F. Bishert Bay, Luck Eveng. Betral. Ann.

- L. Néel, Appl. Sci. Res. B4, 13 (1954).
 F. Rimbert, Rev. Inst. Franç. Pétrole Ann. Combust. Liquides 14, 17, 123 (1959).
 G. Bate, J. Appl. Phys. 33, 2263 (1962).
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Structural Pattern in Central Uplifts of Cryptoexplosion Structures as Typified by Sierra Madera

Abstract. The pattern of deformation in central uplifts of Sierra Madera and other well-known cryptoexplosion structures indicates that inward as well as upward movement of strata formed the uplifts. This kind of movement is incompatible with structures not of impact origin with which they have been compared. The structural style of cryptoexplosion structures, together with features that suggest shock deformation, supports the belief that they are the eroded roots of impact craters.

The debate concerning whether crypto explosion structures (1) are of terrestrial or extraterrestrial orgin began three decades ago (2, 3) and continues. As shown (2), they are roughly circular structures, with a ring depression surrounding a central uplift of disordered and brecciated rocks. The gross structure, as well as the common occurrence of shatter cones (4) and of unusual mineral deformation (5), indicates an origin by explosive release of energy, either from impact (3) or from volcanic gas (2).

Most published explanations of the origin of these structures have been based on inadequate information about their geometry, and give undue weight to the disorder of the central uplifts. Our detailed examination (6) of the Sierra Madera cryptoexplosion structure shows that, despite extensive brecciation and small-scale structural complexity (7), the rocks of the central uplift have a coherent structure and are so arranged that the strata must have moved both inward and upward to

reach their present positions. A similar style of deformation is found in other well-studied cryptoexplosion structures, and casts doubt on a nonimpact origin.

Sierra Madera (8) is a circular body of intensely deformed rocks about 12 km in diameter, 5 km northeast of the Glass Mountains in west Texas. Its intense deformation of rocks is unique in a region where the rocks are otherwise slightly deformed; there is no apparent relation between Sierra Madera and any regional or other local feature. The deformed rocks are Permian limestones and dolomites, like those exposed in a more orderly sequence in the nearby Glass Mountains (Leonard Series, Word, Gilliam, and Tessey formations), and Lower Cretaceous limestones and marls, with a conspicuous basal sandstone (Trinity and Fredericksburg groups, and the lower part of the Washita Group). Records of deep holes drilled for oil and gas show little if any deformation below 2 to 2.5 km under the center of the exposed structure, although the thickness of the