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# Chemomagnetic fields produced by solid combustion reactions

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We report here the observation of *chemomagnetism*, generation of a magnetic field by rapid high-temperature solid reactions producing various oxides. The low-level transient magnetic fields were measured with a high-$T_c$ superconducting quantum interference device magnetometer during the synthesis of both nonferromagnetic and ferromagnetic (ferrite) compounds. The magnetic field was most likely produced by chemoionization processes generated by the moving high-temperature reaction zone. The permanent magnetic field formed by the synthesis of ferromagnetic materials depended on the difference between the combustion temperature and the Curie temperature of the product. © *1999 American Institute of Physics.* [S0003-6951(99)03034-X]

During solid flame combustion a high-temperature front propagates through a solid powder mixture converting it into products.[1,2] We studied the formation of magnetic fields during high-temperature synthesis of two types of reacting solid mixtures. Group 1 was comprised of nonferromagnetic solid reactants yielding nonmagnetic oxides ($BaTiO_3$, $SrTiO_3$, $BaCrO_3/BaCrO_4$, and $TiO_2$). Group 2 consisted of reactants whose ferromagnetic products contained either iron or iron oxides ($Fe/Al_2O_3$, $NaFeO_2$, $BaFe_{12}O_{19}$, and $SrFe_{12}O_{19}$).

The experimental setup is shown schematically in Fig. 1. A quartz tube (1 cm i.d., 6.5 cm long) with a mixture of precursor chemicals (length between 1.0 and 6.5 cm) (a) is placed into a thermally isolated enclosure (b) to protect the high-$T_c$ superconducting quantum interference device (SQUID). The low-level transient magnetic fields are measured by a high-$T_c$ SQUID (Refs. 3 and 4) magnetometer (d), housed in a fiberglass G-10 liquid-nitrogen dewar (e) positioned about 1.0 cm above the sample. Liquid nitrogen maintains the SQUID at 77 K. The sample is ignited with a wooden match (c) through a hole in the heat shield. The SQUID signal is transmitted through a cryogenic cable (f) to outside electronics. High-permeability magnitude shielding (g) eliminates background noise. In some measurements the SQUID signal passes a 5 Hz low-pass filter to decrease ambient noise. The combustion temperature and velocity are determined by thermocouples placed at two positions along the sample.

The temperature front during synthesis of group 1 reactants generated a weak (order of nT), rapidly oscillating magnetic field (Fig. 2). Experimental difficulties prevent simultaneous recording, and hence, comparison of the temperature and magnetic signals. Table I reports the relation between the combustion velocities (in a separate experiment) and magnetic signals. The magnetic signal duration for nonferromagnetic materials were taken as the time between the departure of the signal from its original ''0'' level until it re-

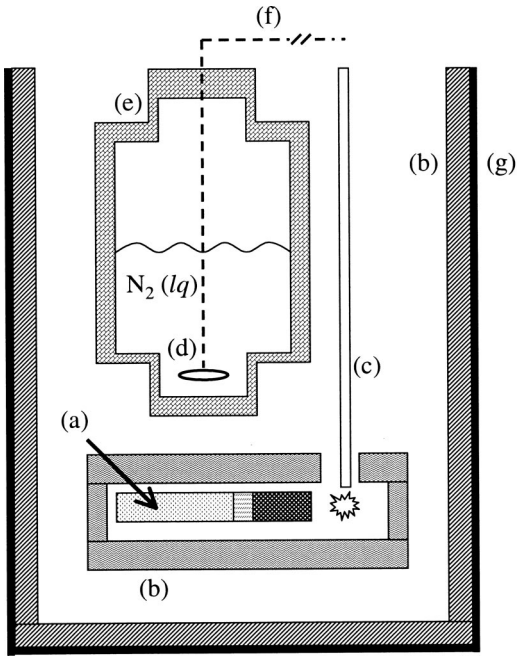


FIG. 1. Experimental setup for chemomagnetic measurements.

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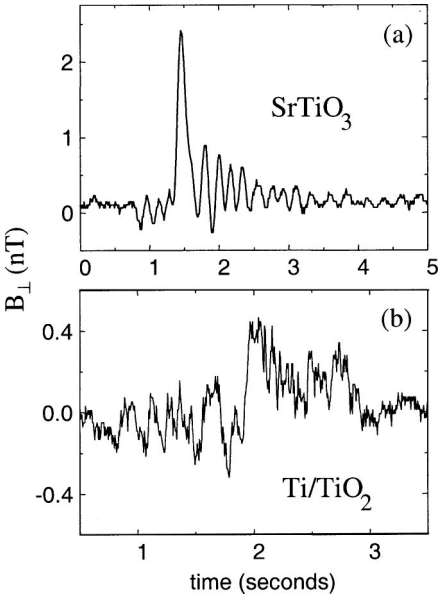


FIG. 2. Magnetic signals of the reactions: (a) $SrO_2+0.4Ti+0.6TiO_2 \rightarrow SrTiO_3$ and (b) $Ti+5.5TiO_2+0.5NaClO_4 \rightarrow TiO_2+0.5NaCl$.

turns (after combustion) to its original level. For ferromagnetic materials, the duration was taken as the time between the initial increase in field magnitude until it stabilized to its final, constant value. For highly exothermic reactions (formation of either $BaTiO_3$, $SrTiO_3$, $SrSiO_3$ with calculated adiabatic temperature exceeding 3000 °C), the fast moving front generated a relatively large magnetic field (with a large peak), followed by a small amplitude, postcombustion, decaying oscillatory magnetic field [Fig. 2(a)]. This magnetic signal usually persisted after the high-temperature front exited the sample (Table I), indicating that postcombustion processes may generate a magnetic field. For mildly exothermic reactions (such as a highly diluted Ti) the magnetic signal included many high-frequency oscillations [Fig. 2(b)]. The corresponding duration of the magnetic signal is close to that of the front movement in the sample. A magnetic signal was formed during the oxidation of Ti by $NaClO_4$ only while the temperature front was within the sample. Since neither the reactants nor products of the group 1 mixtures were ferromagnetic, the magnetic field disappeared after completion of the combustion and postcombustion processes.

Previous studies[5,6] found that solid combustion produces a time-varying electric field in and around the combustion front. Multiple reactions and ionization processes occur during the formation of complex oxides, such as titanates, ferrites, and chromites.[7,8] The peroxide's (or perchlorite's) decomposition in the preheating (pre-front) zone[7,8] may generate short-lived atomic and anionic oxygen and peroxide ions.[9,10] Ions also form by fuel oxidation (Ti, Si, Al, Fe, or $Cr^{3+}$, to $Cr^{6+}$ in reaction between $Cr_2O_3$ and $BaO_2$) in the combustion zone and by reactions among various intermediates in the postcombustion zone. The observed chemomagnetic fields are, most likely, generated by the intense chemoionization processes[11] during the combustion. Transient local excesses of negatively and positively charged ions (for example, in the prefront and front zones) and their relative motions (caused by different diffusivities) may contribute to the oscillatory behavior of the magnetic field. In addition, velocity fluctuations of the moving reaction front may generate oscillations of the magnetic field.

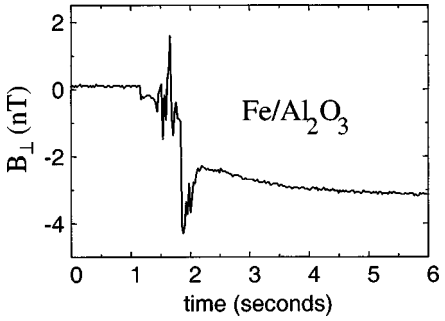


FIG. 3. Magnetic signals of the reaction $Fe_2O_3+2Al \rightarrow 2Fe+Al_2O_3$.

The magnitude of the transient magnetic field produced during the combustion of the second groups of reactants was similar to that created during the synthesis of the nonferrite products. A major difference is that the transient magnetic field formed during SHS of ferromagnetic (ferrite) materials may lead to a permanent magnetic field. For example, SHS of $SrFe_{12}O_{19}$ created a 2000 nT field. The possible occurrence of such an effect was mentioned in Ref. 12. The amplitude of the magnetic signal formed by the termite reaction:

$$Fe_2O_3+2Al \rightarrow 2Fe+Al_2O_3, \quad (1)$$

was twice that of the permanent magnetic field (2.3 nT) generated by the product (Fig. 3). The oscillations lasted for about 1.3 s, roughly equal to the sojourn of the temperature front in the sample, as no postcombustion processes occurred in this case. The magnetic signals observed following the synthesis of the ferromagnetic materials were strongly affected by the difference between the combustion and product Curie temperatures. When the combustion temperature greatly exceeded the Curie temperature, the magnetic field decayed after the combustion wave exited the sample. After

TABLE I. Reactions schemas, maximum oscillation magnitude of magnetic field ($B_\perp$), magnetic signal duration ($t$), front propagating duration ($t_f$), combustion temperature ($T$), and velocity ($U$).

| Reaction schemas | $B_\perp$ (nT) | $t$ (s) | $t_f$ (s) | $T$ (°C) | $U$ (mm/s) |
|---|---|---|---|---|---|
| $BaO_2+0.4Ti+0.6TiO_2 \rightarrow BaTiO_3$ | 6.5 | 1.4 | 0.7 | 3200[a] | 93.3 |
| $SrO_2+0.4Ti+0.6TiO_2 \rightarrow SrTiO_3$ | 2.7 | 4.3 | 1.3 | 3200[a] | 51.0 |
| $SrO_2+0.4Si+0.6SiO_2 \rightarrow SrSiO_3$ | 1.6 | 8.8 | 1.5 | 3100[a] | 43.3 |
| $BaO_2+0.5Cr_2O_3 \rightarrow BaCrO_4/BaCrO_3$ | 0.8 | 6.5 | 6.6 | 755 | 1.8 |
| $Ti+5.5TiO_2+0.5NaClO_4 \rightarrow TiO_2+0.5NaCl$ | 0.7 | 9.5 | 9.5 | 1340 | 3.7 |

[a]Calculated value.

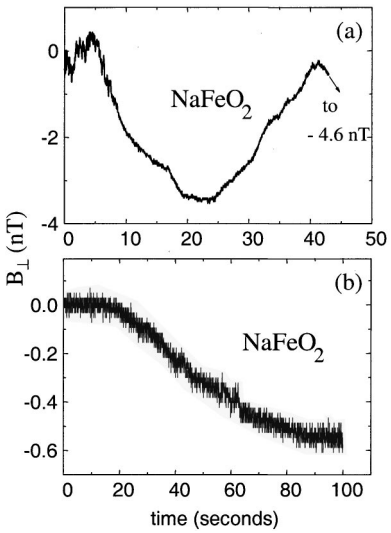


FIG. 4. Magnetic signals of the reactions: (a) $0.5Na_2O_2+Fe\rightarrow 4NaFeO_2$ and (b) $2Na_2O_2+Fe+1.5Fe_2O_3\rightarrow 4NaFeO_2$.

the sample cooled below the Curie temperature, spontaneous magnetization of the product generated a permanent magnetic field, as illustrated in Fig. 4(a) for the reaction

$$Na_2O_2+2Fe\rightarrow 2NaFeO_2. \quad (2)$$

The corresponding combustion and product Curie temperatures were 1210 and 600 °C, respectively.[13] The change in the sign of the measured magnetic field from positive to negative observed in Fig. 4 is not clear at present. A definitive explanation will require more extensive experimental and theoretical modeling studies. A plausible explanation is that the exterior and interior of the sample cool to the Curie temperature at different times and the direction of spontaneous magnetization of these regions may not necessarily be the same. The qualitative features of the magnetic signal were rather different when the combustion temperature was less than the product Curie temperature. For example, when the reactants in reaction (2) were diluted with $Fe_2O_3$, the reaction

$$Na_2O_2+0.5Fe+0.75Fe_2O_3\rightarrow 2NaFeO_2, \quad (3)$$

generated a combustion temperature of 560 °C, which is below the product Curie temperature of 600 °C. The magnetic field in this case monotonically increased to its final value [Fig. 4(b)]. Qualitatively similar magnetic signals were observed during the synthesis of the hard ferrites $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$.

One should not attempt at correlate the magnetic field magnitudes with the combustion temperatures and/or velocities of different systems as other factors may affect it, such as the specific reaction mechanisms, solid mixture compositions, etc. A meaningful comparison must be carried out for the *same* system, similar to that done for sodium ferrite. We are conducting such a study.

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