Magnetocaloric effects in antiferromagnetic iron carbonate magnetized by a pulsed field

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Experimental and theoretical investigations are made of the properties of a phase transition in the field, occurring under adiabatic conditions. It is shown that with increase of the magnetic field, the system moves in phase space along a line of phase transition of first order. In the vicinity of the tricritical point, the adiabaticity is disturbed, because there is an abrupt increase of irreversible absorption of energy from the low-frequency external magnetic field. Thus it is possible to explain from a single point of view the reversible and irreversible processes observed in magneto-optic and magnetocaloric experiments on siderite. The results are also in complete agreement with published data of earlier magnetic measurements.

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1. INTRODUCTION

Investigations of antiferromagnetic crystals in strong magnetic fields are usually made by the pulse method. The pulsed field, changing comparatively slowly (frequency $\Omega \sim 10^4$ s^{-1}), in quasistatic, and therefore the processes occurring in a thermally isolated magnet are adiabatic. In the present paper, a study is made of the properties of phase transitions in an antiferromagnet under adiabatic conditions, and it is also shown that in certain cases the indicated adiabaticity may be disturbed; this in turn must lead to new nonstationary effects. Thus possibilities are opened up for obtaining, by the pulse method, information not only about the static but also about certain dynamic characteristics of magnets.

The behavior of crystals of siderite (iron carbonate, $FeCO_3$) was studied experimentally in pulsed magnetic fields of intensity up to 270 kOe. This uniaxial antiferromagnet, at a temperature $T < T_c$, undergoes in a strong longitudinal magnetic field a transition from the collinear antiferromagnetic phase to a saturated paramagnetic.[1-5] Despite the absence of a spin-flop phase, the transition extends over a field interval of the order of 30 kOe. The occurrence of this interval was explained earlier either by the origination of a large number of sublattices[4] or by quantum effects.[6-10] In the same field interval, a strong magnetocaloric effect[11] and an anomalously large hysteresis of the magnetization curve[4] are observed. The present experimental and theoretical investigations are devoted to explanation of the nature and interrelation of all these phenomena.

In the second part of the paper, a theory of phase transitions in pulsed fields is presented. By regarding magnetization in a pulsed field as an adiabatic process and assuming $dH_c(T)/dT > 0$, where $H_c(T)$ is a line of a first order phase transition, one can show that in consequence of the magnetocaloric effect, a metamagnetic system, with increase of the field, moves in phase space along a line $H = H_c(T)$. On approach to the tricritical point (H_λ, T_λ), where the first-order transition changes to a second-order transition, the process ceases to be adiabatic, because, as was shown in Ref. 12, in the vicinity of the point (H_λ, T_λ) there is an abrupt increase of the irreversible absorption of energy from the low-frequency external field.

The third part describes briefly the method used in magneto-optic and magnetocaloric experiments in strong pulsed fields. The results of the experimental investigations are presented in the fourth section of the paper. It is unambiguously demonstrated that under certain conditions, the phase transition in strong magnetic fields is accompanied by an anomalously large absorption of the energy of the pulsed field; over a single pulse of the field, the heating of the specimen, depending on its initial temperature, amounted to 10–20 K. Heating of the specimen took place both on the forward edge and on the rear edge of the pulse.

In the fifth part of the paper, the reversible and irreversible processes observed in the magneto-optical and magnetocaloric experiments on siderite are explained from a single point of view. In particular, analysis of the results has enabled us to construct a phase diagram of siderite, and also to describe quite completely and consistently the known magnetic measurements.[1-5]

2. PROPERTIES OF PHASE TRANSITIONS IN PULSED FIELDS

1. A magnetic first-order phase transition under adiabatic conditions is accompanied by a caloric effect, i.e., a change of the temperature of the system because of the heat of transition. If the increment of volume of the new phase is dV_2, then the corresponding change of temperature dT can be expressed by a formula of the Clapeyron-Clausius type:

$$dT = \frac{T}{C(T, V_2)} \Delta m(T) \frac{dH_c(T)}{dT} \frac{dV_2}{V}; \quad V = V_1 + V_2. \tag{1}$$

Here $\Delta m(T)$ is the discontinuity of magnetization on the line $H = H_c(T)$; $C(T, V_2) = C_1 + (C_2 - C_1)V_2/V$; C_1 and C_2 are the specific heats and V_1 and V_2 the volumes of the first and second phases. If we suppose that $dH_c/dT > 0$, then the caloric effect is positive.

The change of external field dH necessary in order that the system may remain on the phase-transition line H

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$=H_c(T)$ during an increase of temperature by the amount dT is

$$dH=\frac{dH_c}{dT}dT=\frac{T_c(H)}{C(H,V_2)}\Delta m(H)\left(\frac{dH_c}{dT}\right)^2\frac{dV_2}{V}. \quad (2)$$

Formulas (1) and (2) show that under adiabatic conditions, the phase transition extends over a certain interval of fields and temperatures. But the situation usually considered is one in which the total change of field is small ($\Delta H\equiv H_2-H_1\ll H_1$). Then ΔH is neglected, and the variables that occur in formula (1) are considered independent of T; as a result, the relation (1) is easily integrated:

$$\Delta T=T_2-T_1=T_1\frac{\Delta m}{C}\frac{dH_c}{dT};\quad \Delta H=0. \quad (3)$$

Hence is obtained the well-known conclusion that under adiabatic conditions, a phase transition of first-order in the field is accompanied by the discontinuity of temperature that is often observed in experiments. It is quite obvious that formula (3) is approximate and is valid only under the assumptions made above. In general, the transition under consideration can occur only on change of the field H. We are interested specifically in this general case, in which formula (3) is in general invalid, but (1) and (2) yield the following integral relations (for simplicity, we suppose that $C_1\sim C_2$):

$$\int_{T_1}^{T_2}\frac{C(T)dT}{T\Delta m(T)dH_c/dT}=\int_{H_1}^{H_2}\frac{C(H)(dT_c/dH)^2dH}{T_c(H)\Delta m(H)}=\frac{V_2}{V}, \quad (4)$$

where $H_{1,2}=H_c(T_{1,2})$.

The expression (4) shows that under adiabatic conditions a first-order phase transition extends over a temperature interval from T_1 to T_2 and a field interval from H_1 to H_2. When $V_2=V$, the transition is complete; the corresponding values of T_2 and H_2 are found from equations (4). We note that the upper limit for T_2 (and consequently also for H_2) can be either the tricritical point T_λ or a point at which $H_c(T)$ reaches a maximum ($dH_c/dT=0$). In the first case the singularities of $C(T)$ and $\Delta m(T)$ at $T\to T_\lambda$ may be integrable, but when $T\geq T_\lambda$ we have $\Delta m\equiv 0$. The process described is adiabatic and, of course, reversible.

2. The reversible change of temperature described above and dependent on the transition heat may lead to the result that a first-order transition occurring under conditions of thermal isolation, is completed at the tricritical point (H_λ, T_λ). On the other hand, it has been shown[12] that in the vicinity of a tricritical point, the absorption of energy from a slowly changing field becomes anomalously large. For this purpose, longitudinal pumping in an antiferromagnet was considered in a situation in which the external longitudinal field was divided into two components; a strong constant one and a weak slowly varying one. It is quite obvious that this division is not one of principle. It simplified the calculations, but it should not affect the conclusion: in the vicinity of a second-order pulse transition in a strong, slowly varying magnetic field, there occurs an anomalous absorption of energy, which becomes noticeable after a time of the order of a single period of this field.

The power Q_0 thus absorbed is proportional to $(h_0\omega)^2$, where h_0 is the amplitude of the alternating magnetic pumping field and where ω is the frequency. Therefore by decreasing the frequency and increasing the amplitude of the field, one can leave the value of Q_0 at its previous level. In the case of absorption at a high frequency ($\omega\sim 10^9$–10^{10} s^{-1}), characteristic values of the field amplitude amount to $h_0\sim 0.1$–1 Oe. In the low-frequency case ($\omega\sim 10^4$–10^5 s^{-1}), the field amplitude may reach values $h_0\sim 10^4$–10^5 Oe. Consequently the time τ necessary in order that the irreversible change of temperature of the system

$$\Delta T_r=Q_0\tau/C \quad (5)$$

may become noticeable can be the same in both cases. To judge from the high-frequency experiments, characteristic values are $\tau\sim 10^{-4}$ s, which corresponds to a single period of the low-frequency field.

The change of temperature of the specimen caused by this absorption is

$$\Delta T_r=G\frac{g^6h_0^2R^2}{CT}\frac{\omega}{\gamma}, \quad (6)$$

where g is the gyromagnetic ratio, γ is the attenuation, R is a characteristic dimension of the specimen, and

$$G=4\pi N_A\mu_B^6/\hbar^2a^6c^2k, \quad (7)$$

N_A is Avogadro's number, μ_B is the Bohr magneton, c is the velocity of light, a is the interatomic distance, and k is Boltzmann's constant.

We note that when we are considering absorption in a strong pulsed field H, we may suppose that $h_0/H=\text{const}\ll 1$. Hence it follows that ΔT_r decreases with decrease of the field H. Accordingly, the effective frequency ω_{eff} that must be substituted in formula (6) is proportional to the value of dH/dT at the transition point.

Formula (6) was obtained by consideration of a simple model of the problem, and the absorption described by it is due to nonuniformity of the field in an electromagnetic wave. It must be emphasized that the nonuniformity of the field in an actual solenoid, resulting from finiteness of its dimensions, is as a rule considerably larger than that which we have taken into account, and the absorption may be still greater. Furthermore, a contribution to the absorption may be made by interactions not taken into account in the earlier paper.[12]

3. EXPERIMENTAL METHOD

The investigations of the properties of phase transitions in pulsed fields were made on specimens of the mineral siderite, $FeCO_3$, which had the form of rectangular parallelepipeds with characteristic linear dimensions from one to four millimeters. The pulsed magnetic field, of intensity up to 270 kOe, was produced in a miniature solenoid. The pulse duration was 2 ms. The working volume of the solenoid was a cylinder of diameter 5.5 mm and length 20 mm.

The temperature of the specimen was measured by two methods: magneto-optic,[11] which made it possible to follow the "instantaneous" value of the temperature, and

the usual thermocouple method. The magneto-optic method was based on observation of a group of absorption bands of light in the frequency region 24 000 cm^{-1}. Photographs of the spectrum were made during short (~20 μs) flashes of light, synchronized with one or another section of the magnetic-field pulse. The intensity of the indicated bands can be related to the temperature of the specimen. This inertialess thermometer made it possible to record the temperature of the crystal, if it exceeded 21 K, with accuracy ±0.5 K for an arbitrary value of the varying magnetic field. The investigations were made on a spectrograph DFS-8, with linear dispersion 3 Å/mm. The specimen was thermally insulated from the internal walls of the solenoid with packing, and from the cooling liquid with quartz rods. Specially conducted experiments showed that transfer of heat from the solenoid to the specimen began 0.5 s after the field pulse.

Caloric effects in the present work were also recorded by means of a thermocouple. The good qualitative agreement of the results of the spectral and the thermocouple measurements attested to high reliability of the methods of optical spectroscopy. For measurement of the thermocouple temperature, the crystal was placed in an evacuated tube. With such thermal isolation, no transfer of heat from the solenoid to the specimen was observed. Cooling of the specimen to temperatures near the temperature of the bath was insured by a cooling wire, connecting the specimen and the bath. At bath temperature 14 K, the specimen could be cooled to 20 K.

It should be mentioned that specimens of smaller dimensions were used in the magnetic-optic experiments, and therefore it was possible to choose them with a more perfect macrostructure than in the measurements by means of a thermocouple, where more massive specimens were required.

4. EXPERIMENTAL RESULTS

The properties of the siderite crystals were studied at temperatures from 4.2 to 40 K, in pulsed magnetic fields directed along the third-order symmetry axis C_3.

1. The magneto-optic measurements were made at various values of the field amplitudes H_a and of the initial temperatures T_1 of the specimen. For $H_a < H_{c1} \approx 150$ kOe, no changes of temperature were recorded. In the field interval $H_{c1} < H_a < H_{c2} \approx 180$ kOe, the change of temperature of the specimen occurred in a practically reversible manner. And finally, for $H_a \geq H_{c2}$ absorption of the energy of the field was observed; as a result, at the end of the cycle the specimen temperature T_2 was substantially higher than the initial temperature T_1.

Figure 1 shows two curves of variation of the specimen temperature with the magnetic field intensity H. Curve 1 corresponds to measurements with synchronization of the light pulse with various sections of the pulse of the magnetic field, which at its maximum reached values $H_a = 270$ kOe. This is a typical curve for $H_a > H_{c2}$. Curve 2 was obtained with synchroniza-

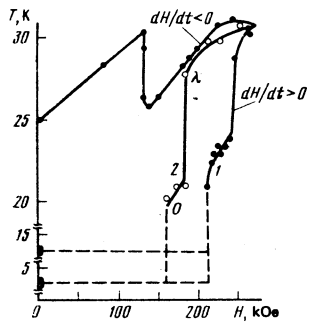

FIG. 1. Variation of values of the magnetocaloric effect with magnetic field intensity at $T_1 = 14$ K: •, $H_a = 270$ kOe; ○, $dH/dt = 0.150$ kOe $\leq H_a \leq 270$ kOe.

tion of the light pulse with pulsed-field maxima $H_a = 150$–270 kOe. It should be mentioned that the position of the experimental points varies somewhat from specimen to specimen (if the crystals were taken from different deposits), but the general form of the curves is retained. Noteworthy features are the discontinuities of temperature not only at the forward edge ($dH/dt > 0$) but also at the rear edge ($dH/dt < 0$) of the pulse, and the lagging effect.

The temperature of the cooling bath in these experiments was 4.2, 14, and 20.4 K. But for $H_a > H$ there was practically no dependence on these initial temperatures. At the same time, for $T_1 \geq 30$ K the magnitude of the magnetocaloric effect began to decrease with increase of T (Fig. 2).

2. The magneto-optical measurements enabled us not only to follow the temperature but also to determine the phase in which the specimen was at prescribed values of the field and temperature. For this purpose, the magnitude $|l|$ of the antiferromagnetism vector was monitored simultaneously with the caloric measurements. This monitoring was accomplished on the basis of the magnitude of the splitting of the exciton lines $\nu_{exc} = 25\,168$ cm^{-1} in the light-absorption spectrum.[13] The presence of the splitting indicated that $l \neq 0$. When $l = 0$ (in the saturated paramagnetic phase), the splitting is absent.

A typical variation of the splitting of the exciton line with the magnetic field intensity is shown in Fig. 3. The measurements showed that at low temperatures T_1, a transition from the antiferromagnetic phase to the saturated paramagnetic actually occurs within the

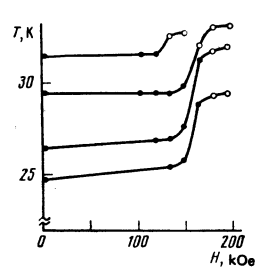

FIG. 2. Variation of values of the magnetoclaoric effect with magnetic field intensity for various values of the initial temperture T_1. •, antiferromagnetic phase; ○, paramagnetic phase.

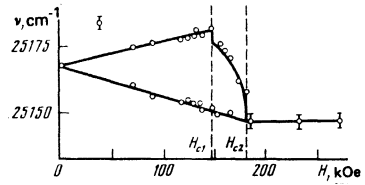

FIG. 3. Variation of splitting of the exciton line 25 168 cm^{-1} with magnetic field intensity: T_1 = 14 K (dH/dt = 0).

field interval 150–180 kOe (H_{c1}–H_{c2}). In the temperature interval 21–25 K, the exact position of the discontinuity in the splitting of the exciton line 25 168 cm^{-1} (i.e., the value of H_{c1}) cannot be determined unambiguously, because of broadening of its components. In the range 25 K < T_1 < 29 K, the whole transition is accomplished near $H_{c2} \approx 180$ kOe, within a field interval not exceeding 10 kOe. On increase of T_1 above 29 K, the field for transition to the paramagnetic phase itself begins also to decrease, as is recorded in Fig. 2.

In a field with amplitude $H_a > H_{c2}$ at $T_1 \leq 20.4$ K, significantly different types of phase transition are traced on the forward and on the rear edges of the magnetic-field pulse; the transition from the paramagnetic phase to the antiferromagnetic (i.e., when $dH/dt < 0$) occurs directly, bypassing the intermediate-state phase that occurs on the forward edge of the field pulse in the interval H_{c1}–H_{c2}.

At large values of dH/dt (above 10^8 Oe/s), hysteresis of the phase transition is observed.[1)] This enabled us to estimate the relaxation time of the magnetically ordered system we are considering. The value of the relaxation time was found to be $\tau_r \sim 10^{-4}$ s. The same order of magnitude for τ_r was obtained by Dudko, Eremenko, and Fridman.[4]

3. As has already been mentioned above, during magnetization of a siderite crystal there occurs a significant change of its temperature. Figure 4 shows these temperature effects, recorded by the thermocouple method. The time $t = 0$ (Fig. 4b) corresponds to application of the magnetic field. The duration of the field pulse is of course so small on the scale of the figure that the beginning and the end of the pulse cannot be resolved. The forward edge of the curves ($dT/dt > 0$) corresponds to transfer of heat from the crystal to the thermocouple, the rear edge ($dT/dt < 0$) to cooling of the crystal-thermocouple system. As is seen from the figure, heating occurs only for initial specimen temperatures $T_1 < T_N \approx 36$ K. One notices the linear section of the curve for $T_1 < T_\lambda \approx 30$ K.

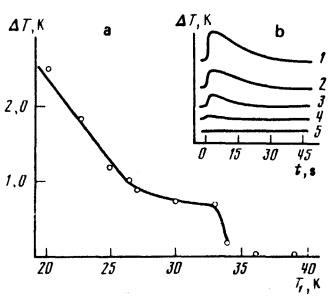

FIG. 4. a, Temperature jump recorded by thermocouple after a pulse of field H_a = 190 kOe, as a function of initial temperature T_1. b, variation of thermocouple reading with time after a pulse of field H_a = 190 kOe, for various values of T_1: 1, 22.5 K; 2, 25 K; 3, 33K; 4, 34 K; 5, 39 K.

The variation of the temperature jump with the amplitude H_a of the pulsed field, for a fixed value of T_1, is plotted in Fig. 5. When the field H is oriented along the axis C_2, there is no magnetocaloric effect (this is indicated both by the magneto-optic measurements and by the measurements with the thermocouple).

5. DISCUSSION OF RESULTS

1. As our measurements show and is known from earlier papers,[1-5,11] at sufficiently low temperatures ($T_1 < 20$ K) anomalies of various physical characteristics of siderite occur at field $H = H_{c1}$. The natural assumption that these anomalies are due to a first-order phase transition occurring under adiabatic conditions, enables us to explain, on the basis of the theory presented, the whole set of phenomena observed in siderite.

From this point of view, the variation of specimen temperature with field (curve $O\lambda$ in Fig. 1) is none other than the line of first-order phase transitions in the HT diagram of siderite. In fact, within the field interval (H_{c1}, H_{c2}) a reversible change of temperature with field was observed. The appearance of irreversible absorption of field energy for $H_a \geq H_{c2}$ indicated that at field H_{c2} the first-order transition changes to a transition of second order; that is, $H_{c2} = H_\lambda$. In accordance with theory, the position of the experimental points on this curve is independent of the temperature T_1; for $H_a > H_{c2}$ and $T_1 < T_\lambda$, the phase transition in the system considered always terminates at the point (H_λ, T_λ), independently of the initial temperature T_1. This means that for this phase transition, within the temperature interval from T_1 to $T_2 \leq T_\lambda$, the whole volume of the specimen does not transform to the new phase. By using formula (4) and the values of the parameters of siderite ($\Delta m \approx 1$ kOe,[4] $dH_c/dT = \Delta H/\Delta T = 1$ kOe/deg, $\Delta H = H_{c2} - H_{c1}$, $\Delta T = T_2 - T_1$, $T_1 = 4.2$ K, $T_2 = 25$ K, $C \approx 10^7$ erg/mol deg), we make a numerical estimate of the value of V_2/V. The result is $V_2/V \approx 0.2$. The rest of the volume transforms to the new phase in the immediate vicinity of T_λ.

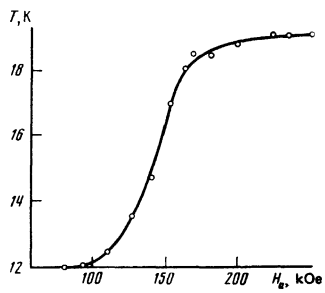

FIG. 5. Temperature recorded by thermocouple as a function of amplitude H_a of pulsed field: T_1 = 12 K.

According to the theory, for $T_1 > T_\lambda$ irreversible absorption of the energy of the pulsed field should be observed. As is seen from Fig. 2, such absorption occurred; with increase of the temperature T_1, the transition field H_c decreased, and with it the absorption. As $T_1 \to T_N$, the field $H_c \to 0$ and the absorption disappeared. Thus the line of second-order phase transitions was recovered, and the phase diagram of siderite has the form shown in Fig. 6.

In conclusion, we shall make an estimate of the change of temperature ΔT_r produced by irreversible absorption of magnetic-field energy at the point (H_λ, T_λ). Substituting in the expression (6) the values of the parameters $(T_\lambda \approx 30$ K, $g \approx 5)$ and regarding the external pulsed field as the sum of two fields, a constant $H = 180$ kOe and an alternating with amplitude $h_0 = 10$ kOe and frequency $\omega = 10^5$ s^{-1} $(\gamma = 10^{-1}\ \omega, R = 1$ cm), we get $\Delta T_r \approx 10$ K. This result agrees well with the experimental data.

2. The plotted phase diagram enables us completely to explain the effects registered by the thermocouple. It is evident from Fig. 4 that absorption of the energy of a pulsed field with amplitude $H_a > H_{c2}$ occurs only when $T_1 < T_N$; in the region of first-order phase transition, i.e., $T_1 < T_\lambda$, the value of the caloric effect ΔT is linear in T_1 and is

$$\Delta T = A(T_\lambda - T_1) + B\Delta T_r(T_\lambda), \tag{8}$$

where A and B are constants describing the properties of the thermocouple. The first term corresponds to a reversible magnetocaloric effect on a first-order phase transition line. The second term is irreversible absorption in the vicinity of the tricritical point (H_λ, T_λ) and is independent of T_1.

When $T_\lambda < T_1 < T_N$, the change of temperature ΔT varies nonlinearly with T_1 and is

$$\Delta T = B\Delta T_r(T_1).$$

With increase of T_1, the value of ΔT decreases, as it should. The results shown in Fig. 5, as was to be expected, agrees qualitatively with the magneto-optic measurements.

3. The temperature jump on the rear edge of the field pulse (Fig. 1) deserves special attention. This most clearly demonstrated the properties of phase transitions in pulsed fields. In this case we have a complete picture of all the phenomena described. The initial temperature of the system is below the tricritical; the amplitude value of the field is $H_a > H_\lambda$. When the field H reaches the value H_{c1}, a phase transition of first order begins (here the effect of a lag showed up). Irreversible absorption of field energy, occurring primarily in the vicinity of (H_λ, T_λ), leads to hysteresis; and as a result, on the rear edge of the field pulse the system falls on the line of phase transition of second order, where the temperature jump indicated above also occurs.

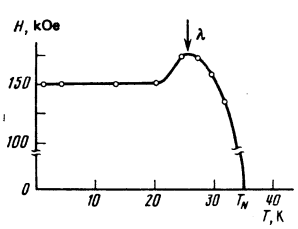

FIG. 16. H-T phase diagram of siderite.

4. Thus it is possible to explain from a single point of view the reversible and irreversible processes observed in the magneto-optic and magnetocaloric experiments on siderite. The results obtained are also in complete agreement with published data of magnetic measurements made earlier.[1-5] First of all we note that according to the plotted phase diagram (Fig. 6), the magnetization curve of siderate should have precisely the form that was obtained in the cited works. In fact, when $T_1 < 20$ K, the phase transition extends in a pulsed field over the interval from H_{c1} to H_{c2}, and the specific magnetic moment increases with the degree of transition of the material from the antiferromagnetic phase to the saturated paramagnetic. In the vicinity of the field H_{c1}, the change of moment occurs quite rapidly [almost jumpwise, in accordance with the slope of the curve $H = H_c(T)$ on the phase diagram]. For $H_{c1} < H < H_{c2}$, a smoother variation of the moment with the field should be observed. And finally, for $H \approx H_{c2}$ there occurs still another jumpwise change of magnetization, which is due to the fact that in the vicinity of the tricritical point a significant volume of the specimen transforms to the saturated paramagnetic phase.

The explanation of the observed threshold character of the hysteresis[4] when $H_a \geq H_{c2}$ becomes quite natural: breakdown of the adiabaticity in the vicinity of the tricritical point and, as a result, absorption of the energy of the pulsed field.

Still another corroboration of the proposed scheme is the behavior of the magnetization and of the susceptibility on the reverse path of the pulsed field. For $T_1 < T_\lambda$ these characteristics should not depend on the initial temperature T_1, as has been mentioned also by Belyi, Kharchenko, and Eremenko.[4] While for $dH/dt > 0$ there are two peaks on the susceptibility curve, corresponding to fields H_{c1} and H_{c2}, for $dH/dt < 0$ a single peak is observed, occupying an intermediate position with respect to these critical fields. This difference can be attributed to the fact that for initial temperature $T_1 < T_\lambda$ the demagnetization of the specimen occurs at a significantly different temperature from its magnetization (Fig. 1), namely at $T > T_\lambda$. As a result, the transition from the paramagnetic state to the antiferromagnetic occurs in a section of the phase diagram corresponding to a transition of second order, and consequently in a field $H_c < H_{c2}$.

It should be noted that the violation of adiabaticity, which is so important in the neighborhood of a phase transition of second order, may show up also in more precise measurements outside this neighborhood. Thus it has been shown[4] that in the field interval $H_{c1} < H < H_{c2}$ the differential magnetic susceptibility has singularities (five spikes), dependent on the rate of change of the magnetic field and disappearing for $dH/dt \to 0$.

6. CONCLUSION

The investigations carried out in this paper enable us to draw the conclusion that in phase transitions occurring in an antiferromagnet in a strong magnetic field, two effects can be observed. First, a phase transition of first order extends over a considerable interval of fields and temperatures (because of the adiabaticity of the process); for definite values of the parameters of the system, the transition may terminate in the vicinity of a tricritical point. Second, in transitions of second order the adiabaticity is violated, and in consequence there is observed an absorption of the energy of a slowly varying magnetic field. The absorption occurs because of the fact that on approach to a line of phase transition of second order, the lowest frequency of antiferromagnetic resonance becomes comparable with the characteristic frequencies of the pulsed field.

The proposed treatment has made it possible to describe quite completely the known experiments performed on siderite; nevertheless, we realize that the problem of siderite requires further theoretical and especially experimental investigations. On the other hand, similar phenomena may occur also in other antiferromagnets. It must be emphasized that according to what has been set forth, the pulse technique may be a good method for construction of certain types of phase diagrams of magnetic materials.

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[1)] This hystersis correlates well with the hystersis of the magnetocaloric effect mentioned earlier.

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