Interpretation of the Hopkinson Secondary Maximum in Terms of Parallel Pumping Theory

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Some former measurements are reviewed concerning a secondary maximum occurring at the high-temperature side of the usual Hopkinson peak if a *dc*-magnetic field is superimposed parallel to the *ac*-field. Since, with regard to single-domain particles embedded in a paramagnetic matrix, the measuring signal is proportional to the high-frequency losses, a secondary maximum indicates some kind of resonance phenomenon which obviously fulfills the geometrical conditions of parallel pumping experiments. It is shown that all other aspects of the experimental results also can be explained sufficiently if being interpreted in terms of parallel pumping theory.

I. Introduction

In the course of high-frequency measurements of Hopkinson peaks done in W. Gerlach's Institute in Munich in order to develop a new method of studying phase transitions and determining Curie points, FRAUNBERGER and STIERSTADT [1] first observed a secondary Hopkinson maximum occurring at the high-temperature side of the normal Hopkinson peak if, during the measurement, a *dc*-magnetic field had been superimposed parallel to the *ac*-field. In 1962 FRAUNBERGER [2] summarized those earlier measurements concerning the Hopkinson secondary maximum and showed the latter to be a very suitable indicator of the Curie temperature even with regard to alloys containing at least one ferromagnetic component.

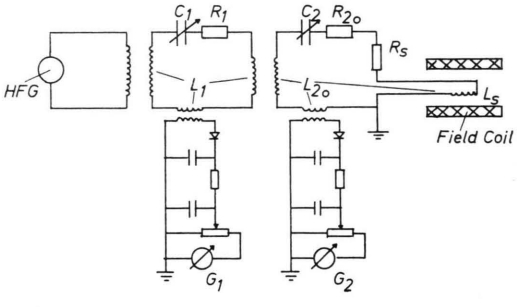

Fig. 1. Schematic sketch of Fraunberger's high-frequency apparatus. HFG designates the high-frequency generator, L_1 the total inductivity of circuit I, C_1 its capacity and R_1 its resistance. L_{20} means the total inductivity of circuit II, exclusive the measuring coil's sample-dependent contribution L_S, whilst R_{20} denotes the total resistivity, again exclusive the sample-dependent contribution R_S. G_1 and G_2 are two galvanometers.

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The apparatus used for these measurements had been described in principle by FRAUNBERGER [3], more in detail by SCHWARZ [4], and recently, with respect to applications in single-domain particle physics, by MARKERT [5]; a schematic sketch is given in Figure 1.

As a typical result some Hopkinson secondary maxima measured above the Curie point of Nickel at increasing *dc*-magnetic fields are shown in Figure 2.

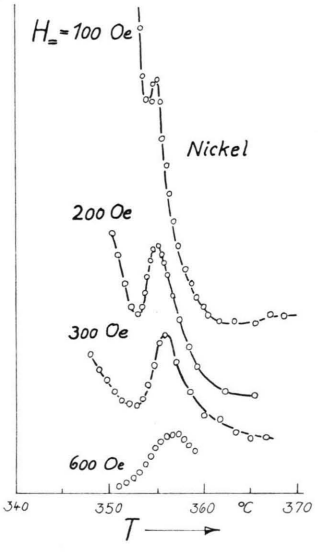

Fig. 2. Hopkinson secondary maxima above the Curie point of Nickel, due to FRAUNBERGER [2].

II. Interpretation in Terms of Parallel Pumping Theory

As far as dealing with the secondary Hopkinson maximum, all of the above cited papers had been confined to phenomenological descriptions of measurements and results. Till now no attempt has been published of understanding the respective measurements by a physical model although, at first view, it seems to be not so difficult to succeed in doing this.

To supplement that interpretation here at least to some extent, let us start from Figure 1. The crucial point is the parallel alignment of both coils producing the *dc*-field and the high-frequency field, respectively. According to SPARKS [6], obviously this is just the geometrical one of the two conditions necessary to establish parallel pumping resonance. The other one requires *ac*-magnetic field amplitudes at least as high as a critical threshold field, given, due to SPARKS [6], by

$$H_c = \hbar\,\omega/4\,\pi\,\gamma\,\mu\,M\,T\,, \qquad (1)$$

where $\hbar = h/2\,\pi$ with h denoting the Planck's constant, and where ω designates the precession frequency which equals half the pump frequency given by the external

[1] K. STIERSTADT, Dissertation, University of Munich 1956.
[2] F. FRAUNBERGER, Z. Angew. Phys. **15**, 474 [1963].
[3] F. FRAUNBERGER, Z. Metallk. **46**, 749 [1955].
[4] O. SCHWARZ, Diplomarbeit, University of Munich 1963.
[5] H. MARKERT, to be published in Geophys. Journ.
[6] M. SPARKS, Ferromagnetic Relaxation Theory, McGraw-Hill Book Company, New York 1964.

ac-field, whilst γ means the gyromagnetic ratio, μ the Bohr magneton, M the change of magnetization associated with the magnons excited by the *ac*-field, and T the relaxation time of these magnons.

Since the order of magnitude of the high-frequency magnetic field amplitude H_m used in the above quoted experiments amounts to about 100 mOe, according to Eq. (1), the second condition is fulfilled if

$$M T \quad 5.1 \cdot 10^{-16}\, \omega / H_m , \tag{2}$$

where again $\omega = \omega_p/2$ with ω_p denoting the pump frequency of the *ac*-field. Substituting M by the shifted susceptibility χ leads to

$$\chi T/(1+N\chi) \gtrsim 5.1 \cdot 10^{-16}\, \omega / H_m^2, \tag{3}$$

with N denoting the demagnetizing factor. Since, in the case of Ni, χ does not exceed an order of magnitude of about 100 emu, condition (3) obviously can be satisfied even at very small values of T if

$$\omega / H_m^2 \lesssim 10^4 \,\mathrm{sec}^{-1} \cdot \mathrm{Oe}^{-2} ,$$

i. e. if one measures in the 10-kilocycle range using field amplitudes of $H_m \gtrsim 1$ Oe. Thus, at least in principle, parallel pumping experiments should be possible by application of Frauenberger's high-frequency apparatus.

Before discussing now further conclusions of Eq. (3), to be drawn with regard to the experimental data given in Fig. 2, let us first ask what kind of result of measurement should be expected when parallel pumping resonance really would take place.

Then, according to SPARKS [6], the spins of each one of the clusters of parallel spins, characterized by their correlation length ξ (see for instance KADANOFF [7], KADANOFF et al. [8], and HALPERIN and HOHENBERG [9]) and by their life time τ, aligned parallel to easy directions, and separated from each other by paramagnetic surroundings, will start to precess around the direction of the effective *dc*-magnetic field by a frequency ω_0 given by

$$\omega_0 = \gamma H_{\mathrm{eff}} \tag{4}$$

and amounting in the case of paralel pumping resonance to half the pump frequency ω_p. But to fulfill the condition

$$\omega_p/2 = \gamma H_{\mathrm{eff}} = \gamma (H_{ex} - N M_1 + H_K) \tag{5}$$

— where H_{ex} designates the external *dc*-magnetic field, N the demagnetizing factor of a cluster of aligned spins, M_1 its resultant magnetization, and

$$H_K = K/2\, M_1 \tag{6}$$

the order of magnitude of the clusters' effective crystalline anisotropy field — by constant ω_p and by arbitrary H_{ex}, obviously H_K is determined by

$$H_K = \omega_p/2\,\gamma + N M_1 - H_{ex} . \tag{7}$$

Thus, because of the temperature dependence of the crystalline anisotropy K and of the clusters' local magnetization M_1, at constant ω_p to any pair of different values H_{ex}' and H_{ex}'' of the *dc*-field there corresponds a pair of different "resonance temperatures" T' and T''. Furthermore, since, according to MARKERT [5], Fraunberger's high-frequency apparatus, if applicated in the usual way, measures the high-frequency losses that become maximum when parallel pumping resonance takes place, we should expect to observe a secondary peak occurring above the usual Hopkinson maximum at a temperature T which, at constant ω_p, should vary with varying H_{ex}.

Reverting now to the experimental results shown in Fig. 2, we state full verification of the above outlined predictions; moreover the temperature T of resonance increases monotonously with increasing H_{ex}! Therefore we think our interpretation in terms of parallel pumping theory to be right although, in the case represented in Fig. 2, the term ω/H_m^2 known from Eq. (3), amounts to about $5 \cdot 10^9$ sec^{-1} Oe^{-2}, thus limiting $\chi T/(1+N\chi)$ to more than $2.5 \cdot 10^{-6}$ sec.

But if it is really possible to do parallel pumping experiments by use of the simple Fraunberger high-frequency apparatus, according to Eqs. (6) and (7), it also should be possible to measure the temperature dependence of the crystalline anisotropy constant, providing that there is a chance to estimate the order of magnitude of the local magnetization M_1 from the scaling laws governing phase transition theory. Although M_1 really seems to be computable as a function of temperature T and of H_{ex}, we shall not discuss this point explicitly here, but just make the following qualitative remarks:

1. Provided that $H_{ex} \gg \omega_p/2\,\gamma$, Eqs. (6) and (7) lead to

$$K \approx 2\, M_1 (N M_1 - H_{ex}) . \tag{8}$$

And because of $\omega_p/2\,\gamma \lesssim 1$ emu, Eq. (8) will hold true if $\omega_p \lesssim 10^7$ sec^{-1}. Thus the right hand side of Eq. (8) will change its sign when, with increasing H_{ex}, according to Fig. 2, the "resonance temperature" T also increases causing by this way a corresponding decrease of M_1! In other words: some degrees above the Curie point of Nickel the crystalline anisotropy constant K is expected to change its sign.

2. At sufficiently high *dc*-fields Eq. (8) can be reduced to

$$-K \approx 2\, M_1 H_{ex} \tag{9}$$

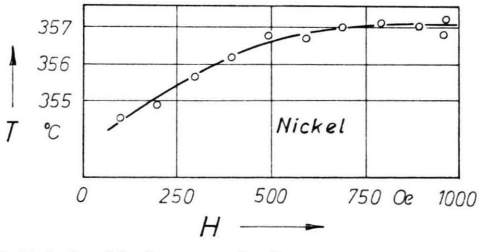

Fig. 3. Relationship between the "resonance temperature" T of the Hopkinson secondary maximum and the *dc*-field amplitude H_{ex} in the case of Nickel, due to FRAUNBERGER [2].

[7] L. P. KADANOFF, Physics **2**, 263 [1966].
[8] L. P. KADANOFF et al., Rev. Mod. Phys. **39**, 395 [1967].
[9] B. I. HALPERIN and P. C. HOHENBERG, Phys. Rev. **177**, 952 [1969].

and hence at a correspondingly high temperature there holds a relationship

$$M_1 \approx -K/2\,H_{ex}\,, \qquad (10)$$

with $K < 0$. Since, according to Fig. 3, at high values of H_{ex} the "resonance temperature" T approaches a limiting value T_1, M_1 as well as K do not longer depend on H_{ex}. Thus, in the case of high *dc*-fields, Eq. (10) describes the approach to saturation magnetization above the Curie point by the same kind of relationship that also holds true with regard to single-domain particles below the critical point.

Acknowledgements

I am very indebted to Prof. Dr. F. FRAUNBERGER by whom I came into contact with the outlined problems.

Elastische und thermoelastische Konstanten des monoklinen Zinndifluorids

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All elastic and thermoelastic constants and the coefficients of thermal expansion of monoclinic SnF_2 are measured. These properties deviate strongly from the behaviour of other difluorides such as CaF_2 and MgF_2. The high anisotropy, the large bulk compressibility and the departures from the Cauchy relations suggest the existence of strong covalent binding components between tin and fluorine atoms. The thermal properties are in accordance with Grünseisen's rule.

Das in der monoklinen Raumgruppe C2/c kristallisierende Zinndifluorid [1], das wegen seiner Isotopenreinheit auch für bestimmte Anwendungen in der Neutronenspektroskopie und für Mössbauer-Experimente geeignet ist, nimmt unter den Fluoriden der zweiwertigen Kationen hinsichtlich seiner Struktur und seiner physikalischen Eigenschaften eine Sonderstellung ein. Mit dem folgenden Beitrag wollen wir auf die Besonderheiten im elastischen und thermoelastischen Verhalten sowie in der thermischen Ausdehnung hinweisen.

Die für die Messungen erforderlichen wasserklaren Einkristalle optischer Qualität mit Abmessungen bis zu 4 cm züchteten wir aus wäßrigen Lösungen sowohl durch Verdampfen als auch durch langsames Senken der Temperatur im Bereich von etwa 50 bis 30 °C. Über die Erfahrungen bei der Züchtung und die Ergebnisse der ersten Messungen an diesen Kristallen berichteten wir schon an anderer Stelle [2].

Die elastischen Konstanten c_{ij} wurden aus den Ausbreitungsgeschwindigkeiten elastischer Wellen in sieben etwa gleichmäßig über die Lagekugel verteilten Richtungen mit Hilfe des von HAUSSÜHL und SIEGERT bei triklinen Kristallen angewandten Verfahrens [3] ermittelt. Für die Messung dieser Schallgeschwindigkeiten benutzten wir das verbesserte Schaefer-Bergmann-Verfahren bei etwa 20 °C (Beugung von Licht an stehenden Ultraschallwellen in dicken Platten bei etwa 15 MHz). Ferner wurden die Temperaturkoeffizienten dieser Wellengeschwindigkeiten aus der Verschiebung der Eigenfrequenzen dicker Platten im Bereich von etwa 20 bis −20 °C gemessen. Für die Berechnung der dreizehn unabhängigen elastischen Konstanten standen insgesamt 16 Gleichungen zur Verfügung. Für die Bestimmung der thermoelastischen Konstanten $T_{ij} = \mathrm{d}\log c_{ij}/\mathrm{d}T$ (T Temperatur) zogen wir die Gleichungen für 20° und die daraus mit Hilfe der bekannten Temperaturkoeffizienten abgeleiteten Gleichungen für 0 °C heran. Beide Gleichungssysteme sind mit nahezu denselben relativen Fehlern behaftet. Die für 20 und 0 °C daraus berechneten elastischen Konstanten besitzen demnach ebenfalls fast dieselben Fehler.

Aus den Differenzen dieser Konstanten für 20 und 0 °C ließen sich dann die thermoelastischen Konstanten wesentlich genauer erfassen als durch direkte Messungen der Wellengeschwindigkeit bei 20 und 0 °C, weil die Verschiebung von Eigenfrequenzen viel genauer gemessen werden konnte als entsprechende Änderungen der Wellengeschwindigkeiten bei Anwendung des Schaefer-Bergmann-Verfahrens.

Zur Bestimmung des vollständigen Tensors der thermischen Ausdehnung haben wir longitudinale Effekte in mehreren Richtungen mittels eines Fizeau-Interferometers zwischen 20 und 0 °C gemessen.

Die Achsen $\boldsymbol{e}_i$ des kartesischen Bezugssystems für den Elastizitätstensor und den Tensor der thermischen Ausdehnung sind gemäß

$$\boldsymbol{e}_1 \parallel \boldsymbol{a}_1\,, \quad \boldsymbol{e}_2 \parallel \boldsymbol{a}_2 \quad \text{und} \quad \boldsymbol{e}_3 = \boldsymbol{e}_1 \times \boldsymbol{e}_2$$

an die kristallographischen Achsen angeschlossen. Diese sind durch das Achsenverhältnis $a_1 : a_2 : a_3 = 1{,}4086 : 1 : 2{,}7291$ und den Winkel $\alpha_2 = 109°$ charakterisiert. In dieser Aufstellung nehmen die wichtigsten Wachstumsformen folgende Indizierung an: {100}, {001}, {111}. Für das spezifische Gewicht bei 20 °C wurde in den Rechnungen der Wert 4,8749 g cm^{-3} eingesetzt.

Für die in Tab. 1 aufgeführten Werte gelten folgende Schranken für die relativen wahrscheinlichen Fehler:

Sonderdruckanforderungen an Prof. Dr. S. HAUSSÜHL, Institut für Kristallographie der Universität zu Köln, *D-5000 Köln*, Zülpicher Straße 49.

[1] G. BERGERHOFF, Acta Cryst. **15**, 509 [1962].

[2] E. ACKER, S. HAUSSÜHL u. K. RECKER,, Vortrag 3. Internationale Konferenz über Kristallwachstum, Marseille 1971. Im Druck in J. Crystal Growth.

[3] S. HAUSSÜHL u. H. SIEGERT, Z. Kristallogr. **129**, 142 [1969].