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Distinctive features of order-order type magnetic transitions due to the exchange-striction mechanism

V. I. Val'kov, É. A. Zavadskiĭ, and B. M. Todris

Donets Physico-Technical Institute, Ukrainian Academy of Sciences

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A technique for computing the P-T phase diagrams of a two-sublattice magnetic substance is developed on the basis of the Bean-Rodbell exchange-striction model. It is shown that, under certain conditions, the Néel temperature T_N can be lower than the temperature at which the energy barrier separating the antiferromagnetic and ferromagnetic phases disappears. In this case the ferromagnetic order should inductively set in near T_N in the sample under investigation. The results of the calculations are confirmed by experiments on the magnetic transitions in manganese arsenide and certain alloys based on this compound. Some new varieties of P-T diagrams of magnetically ordered materials are predicted.

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INTRODUCTION

It has been experimentally established that ferromagnetic ordering in a number of substances is replaced by antiferromagnetic ordering as the temperature is varied.[1–4] Such transitions are usually explained with the aid of Kittel's theoretical model,[5] which is based on the fact that the exchange interaction, linearly varying with the interatomic distance, changes sign at definite, characteristic—for the given substance—dimensions of the crystal lattice. Although Kittel took only one exchange interaction into account in his work, he in fact assumed that there is a second, distance-independent interaction.

Farrel and Meijer[6] took the second coordination sphere directly into account, but considered only the influence of the magnetic field on the equilibrium distances, leaving out of consideration the problems connected with the change of sign of one of the exchange interactions. The calculations in Pal's paper[7] are closest to the real situation, but they were carried out under the assumption that the sublattice magnetization does not change when the ferromagnetic (FM) order is replaced by the antiferromagnetic (AF) order.

As applied to FeRh, which was considered in[7], the above-cited assumption can be considered to be justified. But for a stronger dependence of the exchange interaction on the interatomic distance, the Curie, T_C, or Néel, T_N, temperatures turn out to be different. Then the AF → FM transition temperature can be close to T_N, and the jump occurring in the sublattice magnetization at the time of the transition will be large. This effect has a lesser influence in the inverse FM → AF transition. On the whole then, the $P-T$ phase diagram should change substantially.

In the experimental investigation of the magnetic transitions we encountered in some alloys precisely such a change in the phase diagrams which support the applicability of the above-described approach. Since, in analyzing the results, we found it possible to predict several types of phase diagrams, including FM–AF transitions, it seems to us expedient to generalize the results obtained, which is done in the present paper.

THE THEORETICAL MODEL

The model is based on the assumption that the exchange interaction in both the first, J_1, and second, J_2, coordination spheres depends on the specific volume V. Following Bean and Rodbell's paper,[8] in which an exchange-striction mechanism of phase transitions is developed, we shall assume the dependences $J_1(V)$ and $J_2(V)$ to be linear.

Let the relative disposition of the indicated curves be determined by the parameters indicated in Fig. 1. Let us also introduce the coefficient $n = \partial J_2/\partial V : \partial J_1/\partial V$.

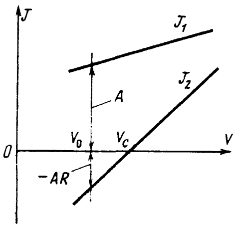

FIG. 1. The dependences, used in the theoretical model, of the exchange interactions on volume. V_0 corresponds to $T=0$; the FM–AF transition is observed in the vicinity of V_c.

Then in the absence of a magnetic field and for the spin number $j=\frac{1}{2}$, the thermodynamic potential of a two-sublattice magnetic substance can be written in the following form:

$$G=-\frac{1}{2}NA\left(1+\beta\frac{\Delta V}{V_0}\right)y^2-\frac{1}{2}NA\left(-R+\beta n\frac{\Delta V}{V_0}\right)y^2\cos\varphi$$
$$-\frac{1}{2K}\left(\frac{\Delta V}{V_0}\right)^2-NkT(S_j+S_{\text{lat}})+P\frac{\Delta V}{V_0}, \qquad (1)$$

where N is the concentration of the magnetic ions; $A=J_1(V_0)=kT_0$; β is a parameter that takes account in the theory of the slope of $J_1(V)$; φ is the angle between the sublattice magnetization vectors; P is the pressure; T is the temperature; K is the compressibility; k is the Boltzmann constant; and y is the relative magnetization of the sublattices.

The first two terms in (1) take account of the exchange energy in the first and second coordination spheres; the third term, the elastic energy that arises when the volume is changed. The contribution of entropy splits into two parts connected with the spin system and the crystal lattice. The last term takes account of the effect of pressure.

In the molecular-field approximation, and for $j=\frac{1}{2}$, the entropy S_j can be written in the following form:

$$S_j=-{}^1\!/_2Nk[(1+y)\ln(1+y)+(1-y)\ln(1-y)-2\ln 2]. \qquad (2)$$

To compute the entropy of the crystal lattice, we can use the relation[9]

$$S_{\text{lat}}=C\ln T+\alpha(V-V_0)/K, \qquad (3)$$

where $\alpha=V^{-1}(\partial V/\partial T)_P$ is the coefficient of thermal expansion. Minimizing (1) with respect to the magnetization, we can derive the temperature dependence of the sublattice magnetization in the implicit form:

$$\frac{T}{T_0}=\frac{(1-R\cos\varphi)y+\beta y\Delta V/V_0}{\operatorname{Ar\,th} y}. \qquad (4)$$

Since $\Delta V/V_0$ in (4) is a function of pressure, the magnetization, and the angle φ, before using this expression for computational purposes, we should determine the equilibrium value of the volume. This can easily be done by solving the equation $\partial G/\partial V=0$. The calculation leads to the following value:

$$\Delta V/V_0={}^1\!/_2AK\beta(1+n\cos\varphi)y^2-PK+\alpha T. \qquad (5)$$

Combining (4) and (5), we obtain

$$\frac{T}{T_0}=\frac{[1-R\cos\varphi-PK\beta(n\cos\varphi+1)]+{}^1\!/_3\eta_0(n\cos\varphi+1)^2y^2}{y^{-1}\operatorname{Ar\,th} y-\alpha\beta T_0(n\cos\varphi+1)}, \qquad (6)$$

where

$$\eta_0={}^3\!/_2NAK\beta^2. \qquad (7)$$

The analysis of (6) enables us to establish the fact that the transitions connected with disordering can be transitions of first or second order. Decisive in this case is the magnitude of the parameter

$$\eta=\eta_0\frac{[1-\alpha\beta T_0(n\cos\varphi+1)](n\cos\varphi+1)^2}{1-R\cos\varphi-PK\beta(n\cos\varphi+1)}. \qquad (8)$$

If $\eta>1$, then the FM–PM and AF–PM transitions (PM represents the paramagnetic phase) are of first order. Second-order transitions will be realized in the case when the inverse inequality obtains.

Let us note some more results that follow from the analysis of (8).

1. The parameters η for the ferromagnetic and antiferromagnetic phases are different. For $n>1$ the value of ε_{FM}, as a rule, significantly exceeds η_{AF}.

2. The characters of the FM–PM and AF–PM transitions are affected differently by pressure. It can easily be verified that pressure increases η_{FM} in a ferromagnet with $\beta>0$, i.e., it facilitates the occurrence of a first-order phase transition. Such an unequivocal conclusion cannot be drawn in the case of an antiferromagnetic material.

3. For the purpose of comparison of the P–T phase diagrams of different magnetically ordered materials, it is convenient to introduce the critical pressures P_{FM} and P_{AF}, which are computed from the condition $\eta=1.0$. The computations lead to the following expressions:

$$P_{\text{FM}}=\{1-R-[1-\alpha\beta T_0(n+1)]\eta_0(n+1)^2\}[K\beta(n+1)]^{-1}, \qquad (9)$$
$$P_{\text{AF}}=\{-1-R+[1+\alpha\beta T_0(n-1)]\eta_0(n-1)^2\}[K\beta(n-1)]^{-1}. \qquad (10)$$

4. Substances for which $\eta=0$ form a special group. In these substances phase transitions of the order-disorder type will be second-order transitions at any pressure, and the transition temperatures should not depend on pressure. It follows from (8) that such a situation can be realized in a ferromagnet with $n=-1$ and in an antiferromagnet with $n=1$. It is possible that the constancy in iron of the Curie temperature in a wide pressure range precisely indicates the fulfillment of the condition $n=-1$.

The main consequence of the marked dependence of the exchange interaction on volume and, hence, of the substantial magnitude of η is the appearance of broad thermal- and pressure-hysteresis regions. This circumstance leads to a situation in which the relation between T_C and T_N is no longer a criterion for stability of one or the other ordered phase. To solve the problem of phase stability, we must resort to minimization of the thermodynamic potential with respect to the angle φ.

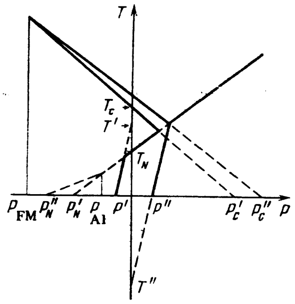

FIG. 2. Generalized $P-T$ phase diagram of a two-sublattice magnetic substance.

The characteristic φ values corresponding to the extremal conditions are found from the equation

$$\frac{\partial G}{\partial \varphi}=\sin\varphi\left(-R+n\beta\frac{\Delta V}{V_0}\right)y^2=0.$$

To the minimum of the energy corresponds only the condition $\sin\varphi=0$. Thus, the model under consideration admits of only collinear solutions: $\varphi=0$ and $\varphi=\pi$. The stability region for these phases is determined by the inequality $\partial^2G/\partial\varphi^2<0$, the FM-AF transition boundaries being themselves determined from the condition $\partial^2G/\partial\varphi^2=0$.

Analysis of (6) and (8) enables us to determine other characteristic points of the phase diagram. We shall, however, omit the computations and tabulate the results. Figure 2 illustrates the conventional symbols used in Table I.

In Fig. 2 all the phase-transition boundaries are depicted as lines. This is somewhat inaccurate, since, in fact, only the boundaries $T_C-P'_C$ and $T_N-P'_N$ are linear. As to the boundaries of the FM→AF transitions in the region $P_{FM}P''_C$ and the AF→PM transitions in the region $P_{AF}P''_N$, they are nonlinear. It should be noted at the same time that the characteristic-point values given in Table I were determined rigorously.

In order to avoid the indicated inaccuracies, we must solve the corresponding self-consistent problems, since the sublattice magnetization is a function of T and P during the transition. In Fig. 3 the dashed lines are lines obtained by the approximate method. The solid lines in the figure correspond to rigorously determined phase-transition boundaries. It follows from Fig. 3 that the hysteresis of the FM→PM transitions gets distorted the most in the approximate (linear) phase diagrams. However, for a qualitative analysis of the magnetic transitions the linear phase diagrams are entirely applicable.

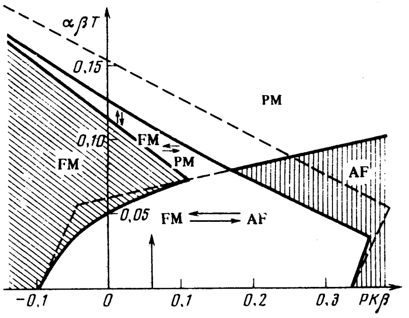

FIG. 3. Calculated phase diagram of a two-sublattice magnetic substance. The diagram was computed with the following values of the parameters: $\eta_0=0.24$, $R=-0.1$, $n=2.68$, and $\alpha\beta T_0=0.075$.

TABLE I. Characteristic points of the $P-T$ phase diagram of two-sublattice magnetic substances.

Conventional symbol	Analytic expression	Conventional symbol	Analytic expression
T_O	$T_0(1-R)[1-\alpha\beta T_0(n+1)]^{-1}$	P'_N	$-(1+R)[K\beta(n-1)]^{-1}$
T_N	$T_0(1+R)[1+\alpha\beta T_0(n-1)]^{-1}$	P''_N	$-[1+R+{}^1/_3\eta_0(n-1)^2]$ $\times[K\beta(n-1)]^{-1}$
P_{FM}	$\{1-R-[1-\alpha\beta T_0(n+1)]\eta_0$ $\times(n+1)^2\}[K\beta(n+1)]^{-1}$	T'	$[R/n+{}^1/_3\eta_0(n-1)][\alpha\beta]^{-1}$
P_{AF}	$\{-1-R+[1+\alpha\beta T_0(n-1)]\eta_0$ $\times(n-1)^2\}[K\beta(n-1)]^{-1}$	T''	$[R/n-{}^1/_3\eta_0(n+1)][\alpha\beta]^{-1}$
P'_C	$(1-R)[K\beta(n+1)]^{-1}$	P'	$[-R/n-{}^1/_3\eta_0(n-1)][K\beta]^{-1}$
P''_C	$[1-R+{}^1/_3\eta_0(n+1)^2]$ $\times[K\beta(n+1)]^{-1}$	P''	$[-R/n+{}^1/_3\eta_0(n+1)][K\beta]^{-1}$

On the face of it, the presence of four parameters in the theory compels us to be somewhat wary. However, for a specific substance we can always find the transition temperatures T_C and T_N at atmospheric pressure, as well as their pressure dependences $\partial T_C/\partial P$ and $\partial T_N/\partial P$. These data are entirely sufficient for the computation of all the parameters of the theory.

The phase diagrams shown in Figs. 2 and 3 correspond to the condition $T'>T_N$. This, undoubtedly, is a particular case. In a real substance the relation between the parameters can be such that the inverse condition $T'>T_N$ will be fulfilled. This, in fact, was assumed in all the early papers. It was assumed that the AF-FM transitions should occur on the line corresponding to the disappearance of the barrier between these states. We especially draw attention to the fact that the Neel temperature can be lower than the temperature at which the energy barrier separating the AF and FM phases disappears. Since the AF phase cannot be maintained above T_N, but $T_C>T_N$, the ferromagnetic order is forcibly established in the sample.

In the following section we shall try to demonstrate that such a situation is realized in some alloys possessing the $B8_1$-type hexagonal crystal lattice.

COMPARISON WITH EXPERIMENT

The question of the applicability of the exchange-striction mechanism to the analysis of magnetic transitions in alloys of the transition metals has in recent years attracted the attention of many investigators. Here the objects of investigation have more often than not been manganese arsenide and alloys based on it, substances in which a first-order FM-PM phase transition is realized even at atmospheric pressure[9,10] and the $P-T$ phase diagram is close to the diagram shown in Fig. 2.[11-13] Only the low-temperature part of the phase diagram remained unstudied, since the carrying out of magnetic measurements under conditions of hydrostatic pressure at low temperatures meets with certain difficulties. Moreover, to determine the AF→FM transition

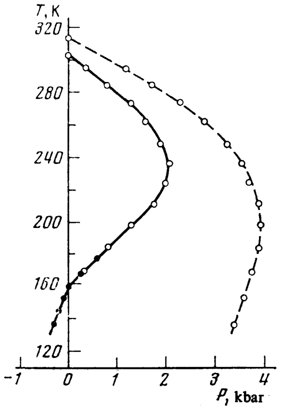

FIG. 4. Experimental phase diagram of manganese arsenide. The continuous curve corresponds to the boundary for spontaneous appearance of the FM phase, while the dashed curve corresponds to the boundary for the disappearance of this phase. o) The results of the direct measurements; ●) the results of the extrapolation of the dependences $H_c(P)$.

boundary, one should carry out the measurements with alternation of the measurement of the temperature and pressure, which has thus far not been done.

We have carried out similar measurements on manganese arsenide and on alloys obtained by the partial substitution of the Mn atoms in MnAs by Ti and Ni atoms. The magnetic properties were studied by the induction method in a high-pressure chamber. In this case in the high-pressure vessel were located: a microsolenoid that allowed the measurements to be carried out in both weak (~ 100 Oe) and strong (up to 270 kOe) pulsed magnetic fields, a system of differentially connected coils, in one of which the sample under investigation was located, and a pressure gauge.

As the pressure-transmitting medium, we used especially pure benzene, which ensured the possibility of carrying out the magnetic measurements right down to a temperature of 140 K in regimes in which the temperatures were fixed and the pressure was continuously varied. The use of an electronically controlled thermostate enabled us to stabilize the temperature to within 0.5 K.

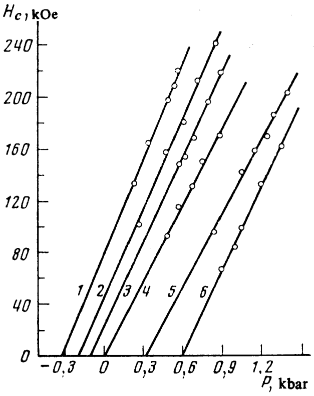

FIG. 5. Variation of the critical field of manganese arsenide under the action of pressure at different temperatures: 1) for $T = 137$ K; 2) for 145 K; 3) for 153 K; 4) for 160 K; 5) for 169 K; 6) for 177 K.

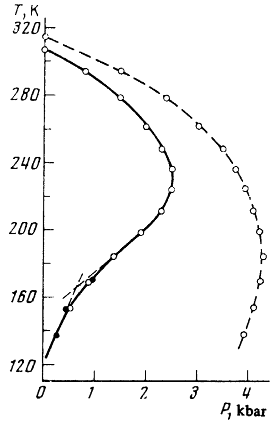

FIG. 6. Experimental phase diagram of the alloy $Mn_{0.97}Ti_{0.03}As$. The conventional symbols are the same as in Fig. 4.

Figure 4 shows the phase diagram of manganese arsenide. The open circles in the figure correspond to the results of the direct measurements; the closed circles, the results obtained by the extrapolation method. The FM phase is always realized inside the region bounded by the continuous curve, while the PM and AF phases are realized outside the region bounded by the dashed curve, the PM phase at $T > T_N$ and the AF phase at $T < T_N$. In the intervening region that phase is maintained which was realized before the operating point moved into this region as a result of a change in temperature or pressure. However, if the initial phase in this region turns out to be the PM or AF phase, then the appearance of the FM phase can be induced under the action of a magnetic field H_c. In Fig. 5 we show the pressure dependence of H_c, measured at different temperatures on one of the investigated MnAs samples.

Analysis of the curves $H_c(P)$ allows us to draw three conclusions.

a) The function $H_c(P)$ turns out to be linear in the entire pressure range.

b) The extrapolation of the linear section of the $H_c(P)$ curve to zero field allows the determination of the pressure at which the FM phase should arise spontaneously.

c) In a comparable pressure range the phase-transition boundaries found by direct measurements and by the extrapolation method agree well with each other.

It appears that the foregoing justifies the above-described extrapolation procedure. We only need to add that this is the only way of obtaining the phase boundaries in the region of negative pressures.

Returning to Fig. 4, let us note that a kink in the AF → FM transition boundary in the phase diagram is clearly visible at 160 K. The fact that to this kink corresponds atmospheric pressure is somewhat of an accident. In Fig. 6 we show the phase diagram for the alloy $Mn_{0.97}Ti_{0.03}As$, whose AF → FM transition boundary

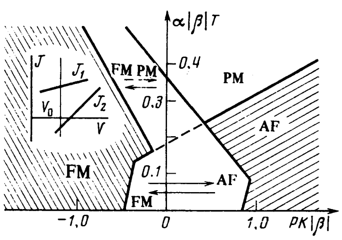

FIG. 7. Calculated phase diagram of the "latent-ferromagnetism" type. The parameters used are: $\eta_0=1$; $R=0.3$; $n=2$; $\alpha\beta T_0=0.1$.

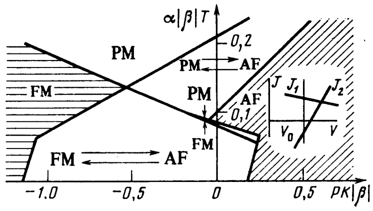

FIG. 8. A new variant of the ***P–T*** phase diagram. The parameters used are: $\eta_0=1$; $R=0.3$; $\beta<0$; $n=-2$; $\alpha|\beta|T_0=0.1$.

also exhibits a kink, but now in the region of positive pressures. Thus, the experiments performed on manganese arsenide and on a number of alloys exhibit in the phase boundary in the P-T diagram a kink which, according to the theoretical model, should be observed in the case when the temperature T' is higher than the Néel temperature.

Comparing Figs. 6 and 4, we can easily notice that the alloyage of manganese arsenide shifts, as it were, the coordinate origin in the phase diagram along the pressure axis. Such a shift becomes comprehensible if we take into account the fact that MnAs and TiAs are isostructural, but that the parameters of the crystal lattice and the volume of the unit cell of titanium arsenide are somewhat larger than the corresponding quantities for manganese arsenide. Such alloying should increase V_0, decrease the parameter R of the theory (see Fig. 1), and shift the phase diagram to the right relative to the coordinate axes. The partial substitution of the As atoms in MnAs by Sb atoms should also lead to this, and indeed it does. The analysis of the results of the study of the magnetic transitions in the alloys $MnAs_{1-x}Sb_x$ allowed us to establish the fact that, at least in the concentration range $0 \leqslant x \leqslant 0.09$, the relation $R=0.10-5x$ is fulfilled.

The effect of alloying with Co, Ni, and Fe on the deformation of the phase diagram of manganese arsenide is opposite to the effect found for Ti. Such alloying decreases V_0 and leads to a shift of the diagram to the left relative to the coordinate axes. Since the kink in the AF → FM phase transition line then shifts toward the region of negative pressures, we do not give these diagrams here.

It would be wrong to overestimate the role of the numerical agreement obtained with the aid of quite a crude model between the experimental and theoretical diagrams. Nevertheless, we can state that the computed—on the basis of the experimental data—parameters of the theory allow us to sufficiently accurately compute the AF → FM transition boundary in the investigated samples.

We should like to note another method of comparing the experimental results with the theoretical results without the use of the parameters of the theory. It follows from Table I that the slope of the low-temperature part of the AF → FM transition boundary should be determined by the relation $\partial T_c/\partial P = K/\alpha$. Using the following data: $K=4.55\times10^{-12}$ cm^2/dyn and $\alpha=5.7\times10^{-5}$ deg^{-1}, which are typical of manganese arsenide,[9] we obtain $\partial T_c/\partial P = 79.5$ deg/kbar. It follows from Fig. 4 that $\partial T_c/\partial P = 70$ deg/kbar. In this agreement should be seen another affirmation of the fact that the theoretical model accurately describes the magnetic transitions observed in real magnetically ordered materials.

CONCLUSION

Thus far we have considered only one particular case of phase diagrams, i.e., the case corresponding to $\beta>0$, $n>1.0$, and $R<0$ (Fig. 4). By varying the parameters of the theory, we can obtain many varieties of phase diagrams. Let us indicate only two of them. Figure 7 shows a calculated phase diagram obtained for a set of parameters that differs from the set used in Fig. 3 primarily in the sign of the parameter R But this led to the result that the FM phase should arise in the region of negative pressures. Such phase diagrams have been experimentally observed.[13,14] If we change the sign of β, then the diagram again undergoes a modification (Fig. 8). Now the AF phase will arise even at atmospheric pressure and will be retained down to absolute zero and the AF–PM transition should become a first-order transition. On the other hand, the FM phase can be induced at low temperatures by a strong magnetic field. This induction will be irreversible.

We can also identify other phase-diagram characteristics connected with other parameters of the theory. Therefore, a comprehensive investigation of the varieties of P–T diagrams theoretically predicted for magnetically ordered materials should, in the final analysis, lead to the use of the exchange-striction mechanism as a method of studying the form of the dependence of the exchange interaction on the interatomic distance.

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Theory of radiation from charged particles channeled in a crystal

M. A. Kumakhov

Nuclear Physics Research Institute of the Moscow State University

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A new physical effect is theoretically predicted, namely intense spontaneous emission of γ photons by relativistic channeled particles. The theory of this phenomenon is developed by the method of quantum electrodynamics and classical mechanics. Both approaches give practically the same results. In the quantum approach, the radiation is due to the transitions between different levels formed in the potential of the atomic planes and chains. At particle energies ~0.1–10 GeV, the radiation is most intense in the band from 0.1 to several dozen MeV. Although the total radiation intensity is less than the bremsstrahlung intensity, the spectral density in the region of the maximum of the radiation can greatly exceed the bremsstrahlung density. The effective cross section for the photoabsorption on the nuclear transitions in this region is larger by several orders of magnitude for the radiation of the channeled particles than for the bremsstrahlung, thus uncovering, in principle, new possibilities in nuclear physics. It is shown than the radiation has several fundamental attributes that distinguish it from the ordinary types. The feasibility of observing this effect in experiment is discussed.

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INTRODUCTION

The channeling effect has been the subject of a large number of investigations (see, e.g., Gemmel's review).[1] In this article we consider mainly channeling of light particles, positrons and electrons. According to Lindhard's predictions[2] it has turned out that with increasing energy the treatment of channeling by classical mechanics becomes more and more correct.[3–5] The angular distributions and the different possible states of the particles were thoroughly measured and calculated.[6–11]

The behavior of relativistic positrons turned out to be close to the behavior of protons, and to explain electron channeling along an atomic chain, the "reseton" model has been proposed.[12] The electron turns out in this case in a bound state in the potential well of the chain and moves around the chain. It was shown experimentally[13] and theoretically[14] that when positrons are channeled the bremsstrahlung is substantially suppressed because the contribution of the short-range collisions decreases. At the same time, the bremsstrahlung increases in the case of electron channeling.[13]

Kalashnikov *et al.*[15] have shown that for nonrelativistic particles there is produced in the channel one level. In this case, naturally, radiation is then possible. A situation is possible, however,[15] in which the wave function of the particle entering the crystal differs from the ground state, so that radiation becomes possible by a transition to the ground state. Even earlier, Thompson[16] had suggested the possibility of stimulated emission by protons in transitions to different states that are formed in the channel. This stimulated emission, in Thompson's opinion, lies in the infrared region and is due to particle scattering by phonons.

In this paper we consider a new type of radiation, which is possible when particles are channeled. It is known[17] that a large number of levels is produced when the energy of the particles in a channel is increased. Spontaneous emission becomes possible, in principle, as a result of transitions between these levels. The relativistic electrons and positrons emit in this case hard γ rays. The possible existence of this effect was proposed in[18]. This effect can also be interpreted on the basis of classical mechanics. The radiation power in the frequency region in which it takes place, turns out to be unusually large, exceeding by approximately two orders of magnitude the bremsstrahlung and by 6–9 orders of magnitude the radiation power of modern synchrotrons.

In the construction of the theory, no account was taken of certain secondary processes, such as dechanneling (see, e.g.,[19–21] on this subject), the effect of the radi-

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