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1993 J. Phys.: Condens. Matter 5 3443

(http://iopscience.iop.org/0953-8984/5/21/007)

J. Phys.: Condens. Matter 5 (1993) 3443–3452. Printed in the UK

Magnetic properties of antiferromagnetic superlattices

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Received 21 July 1992, in final form 10 November 1992

Abstract. We investigate the magnetizations and magnetic susceptibilities as functions of temperature in the presence of external fields, and the H–T phase diagrams of three typical superlattices, the 4_a+4_b, 3_a+4_b and 5_a+5_b superlattices, formed from two completely different antiferromagnetic materials. Because of the competition between the exchange and the Zeeman energies, many different phases can exist in the 3_a+4_b and 5_a+5_b superlattices. The magnetic susceptibility curve, and the magnetization curve in some special cases, can show clearly the transition from a twisted state to another state (except a paramagnetic state). In particular, for the 5_a+5_b superlattice with $J_a = J_I = 0.2J_b$ in an appropriate field, the magnetization curve is discontinuous at the transition point from the antiparallel state 1 to the twisted state.

1. Introduction

Recently, an ionic antiferromagnetic superlattice composed of two different antiferromagnetic slabs, FeF_2 and CoF_2, has been grown on a ZnF_2 substrate [1]. Of course, other ionic antiferromagnetic superlattices can also be prepared by means of the sputtering technique presented in [1] and, generally speaking, these superlattices may be described better theoretically by a localized-spin model. In the last few years, many physicists have presented theoretical studies on the spin-wave spectrum, phase transition, magnetization, etc, of various magnetic superlattices, such as the ferromagnetic superlattices [2], magnetic superlattices composed of identical or different ferromagnetic films with antiferromagnetic coupling at the interfaces [3, 4], and ferromagnetic–antiferromagnetic superlattices [5]. For antiferromagnetic superlattices consisting of two different antiferromagnetic films, Diep [6] has studied theoretically the most simple antiferromagnetic superlattice using the Green function technique and Monte Carlo simulations and has shown the temperature dependence of the sublattice magnetizations and the quantum fluctuation at low temperatures; further, Zhong [7] has studied the spin wave spectrum of an antiferromagnetic superlattice. However, the above studies on antiferromagnetic superlattices all adopted $S_a = S_b$ and $g_a = g_b$ (S and g are the spin quantum number and the effective g-factor, respectively) or the fact that the magnetic moments of different atoms are the same. For antiferromagnetic superlattices with different magnetic moments on different atoms, Xuan-Zhang Wang *et al* [8] have discussed and calculated the longitudinal and transverse susceptibilities in different external fields and have found some interesting properties. At the same time, Carrico and Camley [11] have

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investigated the temperature dependence of the magnetization of the superlattices. In this paper, we shall study other magnetic properties and the phase transitions of this superlattice in various external magnetic fields.

2. Model and formalism

We consider an antiferromagnetic superlattice with a BCC structure and (001) interfaces, and let the period of the chemical structure be given by $P = N_a + N_b$ (N_a and N_b are the numbers of atomic planes in a slab a and a slab b, respectively). The Heisenberg Hamiltonian in the presence of an external field is described by

$$\hat{H} = \sum_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \mu_B \boldsymbol{H} \cdot \sum_i g_i \boldsymbol{S} \tag{1}$$

where $\boldsymbol{H}$ represents an external field and g_i is the effective g-factor of the atom at site i; J_{ij} is the exchange interaction between spins at the nearest-neighbour sites i and j, which equals J_a if both spins lie in slab a, J_b if both spins lie in slab b and J_I if one spin belongs to slab a and the other to slab b.

According to the mean-field approximation, in which many interesting results for the magnetic superlattices are obtained [3, 4], some of which agree with the experimental results [9, 10] qualitatively, for any temperature and external field, the average value of any atomic spin in the ith atomic plane parallel to the interfaces can be written as

$$\langle S_i \rangle = S_i B(y_i) \tag{2}$$

with

$$y_i = (S_i/k_B T)[g_i H \cos\theta_i - 4J_{i-1i}\langle S_{i-1}\rangle \cos(\theta_i - \theta_{i-1}) - 4J_{ii+1}\langle S_{i+1}\rangle \cos(\theta_{i+1} - \theta_i)] \tag{3}$$

where we have taken the Bohr magneton $\mu_B = 1$, $B(y)$ is the Brillouin function and θ_i is the twist angle through which any spin in the ith atomic plane rotates from the direction of the external field. The angle depends on the directions and magnitudes of the spins in the $(i-1)$th atomic plane and the $(i+1)$th atomic plane, as well as the magnitude of the external field. If we have the coordinate system where the X–Z plane is parallel to the interfaces, all spins are in the X–Z plane and the external field is in the direction of the Z axis, θ_i is given by

$$\theta_i = \tan^{-1}[h_x(i)/h_z(i)] \tag{4}$$

where $h_x(i)$ and $h_z(i)$ are the components of the effective field on a spin in the ith atomic plane:

$$h_z(i) = -4J_{ii-1}\langle S_{i-1}\rangle \cos\theta_{i-1} - 4J_{ii+1}\langle S_{i+1}\rangle \cos\theta_{i+1} + g_i H \tag{5}$$

$$h_x(i) = -4J_{ii-1}\langle S_{i-1}\rangle \sin\theta_{i-1} - 4J_{ii+1}\langle S_{i+1}\rangle \sin\theta_{i+1}. \tag{6}$$

In order to avoid divergences in numerical calculations using the computer, we change equations (3) and (4) into, respectively,

$$y_i = (S_i/k_B T)[h_z^2(i) + h_x^2(i)]^{1/2} = (S_i/k_B T)h_i \tag{7}$$

$$h_z(i) \sin\theta_i = h_x(i) \cos\theta_i \tag{8}$$

with an appropriate magnetic period boundary condition. Equations (2) and (8) contain a total of $2P_m$ equations (P_m is the period of the magnetic structure), which constitute a set of simultaneous equations. Using a simulation Newton method to calculate directly the set of simultaneous equations, one can obtain the values of $\langle S_i \rangle$ and θ_i. Therefore the total magnetization of a unit magnetic cell is

$$M = \sum_{i=1}^{P_m} g_i \langle S_i \rangle \cos\theta_i \tag{9}$$

and the average magnetic susceptibility per atom is given approximately by

$$\chi = (1/P_m)(\mathrm{d}M/\mathrm{d}H) \simeq (1/P_m)\{[M(H+\Delta H) - M(H)]/\Delta H\}. \tag{10}$$

Because the set of simultaneous equations can have more than one solution, one should look for the solution corresponding to the state with the lowest free energy. The free energy is given by

$$F = -k_B T \ln Z \tag{11}$$

with the partition function Z [3] of a unit magnetic cell in the superlattice:

$$Z = \prod_i \frac{\sinh[(2S_i+1)h_i/2k_B T]}{\sinh[h_i/2k_B T]}. \tag{12}$$

Applying the theory outlined above, one can determine the H–T phase diagram, the total magnetization and the magnetic susceptibility related to the stable state.

3. Results and discussion

The superlattices considered in this paper are more complex. According to the spin configurations for ground states ($T = 0$ and $H = 0$), there are mainly three kinds of superlattice:

Superlattice (I). The number of atomic planes in slab a and the number of atomic planes in slab b are even, the period P_m of the magnetic structure is equal to the period P of the chemical structure, or $P_m = P = N_a + N_b$, and this superlattice is completely antiferromagnetic.

Superlattice (II). The number of atomic planes in one slab is odd and the number of atomic planes in the other slab is even, $P_m = 2P = 2(N_a + N_b)$, and the superlattice is also antiferromagnetic.

Superlattice (III). The number of atomic planes in slab a and the number of atomic planes in slab b are odd, $P_m = P = N_a + N_b$, and there is a smaller residual magnetization if $S_a g_a \neq S_b g_b$; in other words, the magnetizations of the sublattices in the superlattice do not cancel each other completely.

Because of the differences between the three kinds of superlattice for $T = 0$ and $H = 0$, their phase transitions, magnetizations or magnetic susceptibilities will be very different qualitatively. Considering generality and comparison, in numerical calculations, we take the antiferromagnetic superlattice with $S_a = 2$, $g_a = 2.32$, $S_b = \frac{3}{2}$ and $g_b = 2.00$, and consider two cases:

(1) $J_b = J_I = 0.8J_a$, which implies that the Néel temperature of the bulk material a is twice that of the bulk material b (or $T_{N_b} = \frac{1}{2}T_{N_a}$);

(2) $J_a = J_I = 0.2J_b$, which corresponds to $T_{N_b} = 4.5T_{N_a}$.

We choose three typical superlattices, $4_a + 4_b$, $3_a + 4_b$ and $5_a + 5_b$ superlattices, as the samples. In this paper, the twisted state is one in which the spins in any atomic layer parallel to the interfaces have an angle with respect to the direction of the external field.

The total magnetization and magnetic susceptibility as functions of temperature, and the H–T phase diagram of the $3_a + 4_b$ superlattice are shown for $J_b = J_I = 0.8J_a$ in figure 1, and for $J_a = J_I = 0.2J_b$ in figure 2.

From figure 1, we find that there are four states: a twisted state (TS), state A_1, state A_2 and the paramagnetic state, the spin configurations of which are indicated in figure 3(*a*). From figure 3(*a*), we can see that, in state A_2, all spins in the atomic slabs b of the superlattice are parallel to the direction of the field but, in state A_1, this case does not exist and the spins on the surfaces of the atomic slabs b are antiparallel to the direction of the field. For low temperatures and moderate fields, the state is the twisted state. As the temperature is increased, the state changes from the twisted state to state A_1; at the same time, the period of the magnetic structure changes from $2(3_a + 4_b)$ to $3_a + 4_b$. At the transition point, the magnetization approaches its maximum, and the susceptibility curve for a smaller field has a very sharp peak and is discontinuous. Across the state A_1 region the susceptibility increases with increasing temperature and, in the state A_2 and paramagnetic state regions, it decreases monotonically as the temperature is increased. The transition from the twisted state to state A_1 is accompanied by a rapid change in magnetic susceptibility so that this transition can be easily found in experiments.

From figure 2 with the parameters $J_a = J_I = 0.2J_b$, we see that the only phase transition that can appear is a transition from the twisted state to the paramagnetic state. The spin configurations of the twisted state for two different temperatures are shown in figure 3(*b*). Figures 2(*b*) and 2(*c*) show that the magnetization and susceptibility change continuously in the whole temperature region. In addition, the susceptibility of a high field $H = 0.8J_b$ has its maximum at a finite temperature.

Comparing figure 1 for $J_b = J_I = 0.8J_a$ with figure 2 for $J_a = J_I = 0.2J_b$, one finds that the $3_a + 4_b$ superlattice, for different parameters, can have completely different magnetic behaviours in external magnetic fields, even qualitatively.

The $5_a + 5_b$ superlattice is very interesting and many phases for it can be found for various external fields. Because the magnetizations of the sublattices in the superlattice cannot cancel each other completely, the macroscopic and microscopic properties will be very different qualitatively from those of the other two superlattices. Here we also consider two special cases:

(1) $J_b = J_I = 0.8J_a$;

(2) $J_a = J_I = 0.2J_b$.

The magnetization, magnetic susceptibility and phase diagrams for these cases are given in figures 4 and 5, respectively.

First we discuss case (1). The phase diagram in figure 4(*a*) shows that, depending on the external field and temperature, five states can appear in the superlattice. The states are a twisted state, the antiparallel state where the moments of neighbouring atoms are antiparallel to each other in the direction of the external field, the states labelled B_1 and B_2, and a paramagnetic state. The spin configurations corresponding to these states are illustrated in figure 3(*c*) where the difference between states B_1 and B_2 is similar to that between states A_1 and A_2 in figure 3(*a*). Figures 4(*b*) and 4(*c*) show the temperature dependences of the

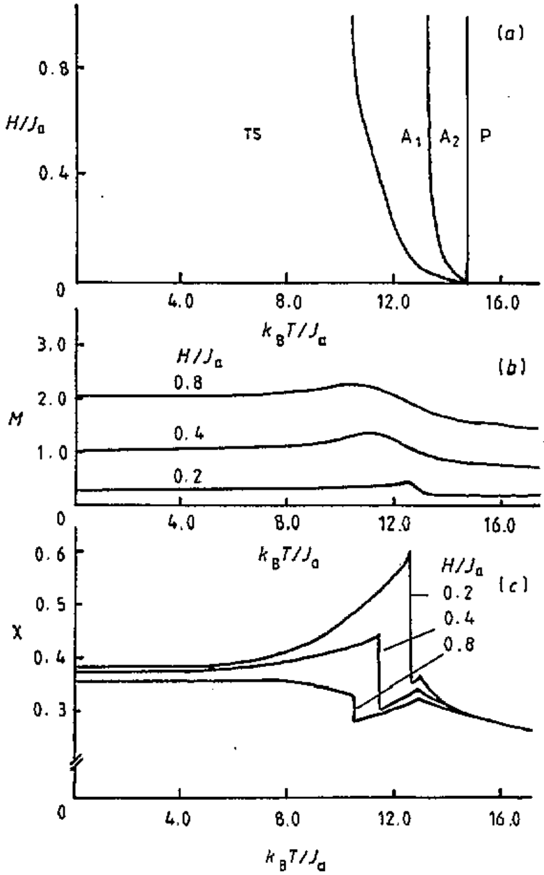

Figure 1. Properties of a $3_a + 4_b$ superlattice for $J_b = J_I = 0.8J_a$: (*a*) the *H*–*T* phase diagram in which the twisted state (TS), A_1, A_2 and the paramagnetic state (P) represent the stable states of the superlattice; (*b*) the magnetization *M* versus temperature for different external fields; (*c*) the magnetic susceptibility as a function of temperature for different external fields.

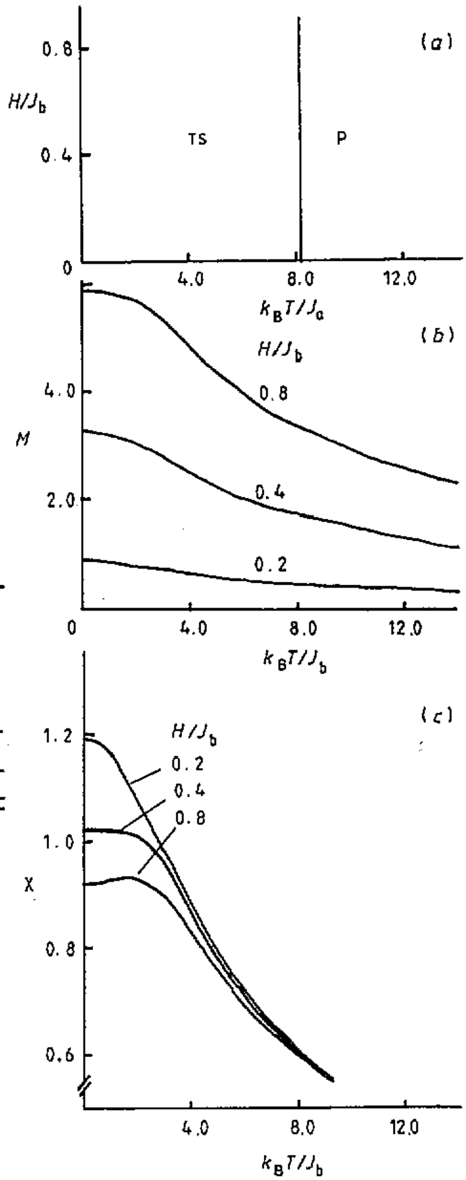

Figure 2. Results for the $3_a + 4_b$ superlattice for $J_a = J_I = 0.2J_b$. (*a*)–(*c*) are as in figure 1. Only two states, the twisted state and the paramagnetic state, can be found in the superlattice. The magnetization and magnetic susceptibility curves change continuously with temperature.

magnetization and magnetic susceptibility, respectively. As is easily seen, the magnetization and susceptibility in the twisted state are insensitive to the temperature or do not change approximately with the temperature. The transition point from the twisted state to the antiparallel state is reflected obviously by the magnetization and susceptibility as functions

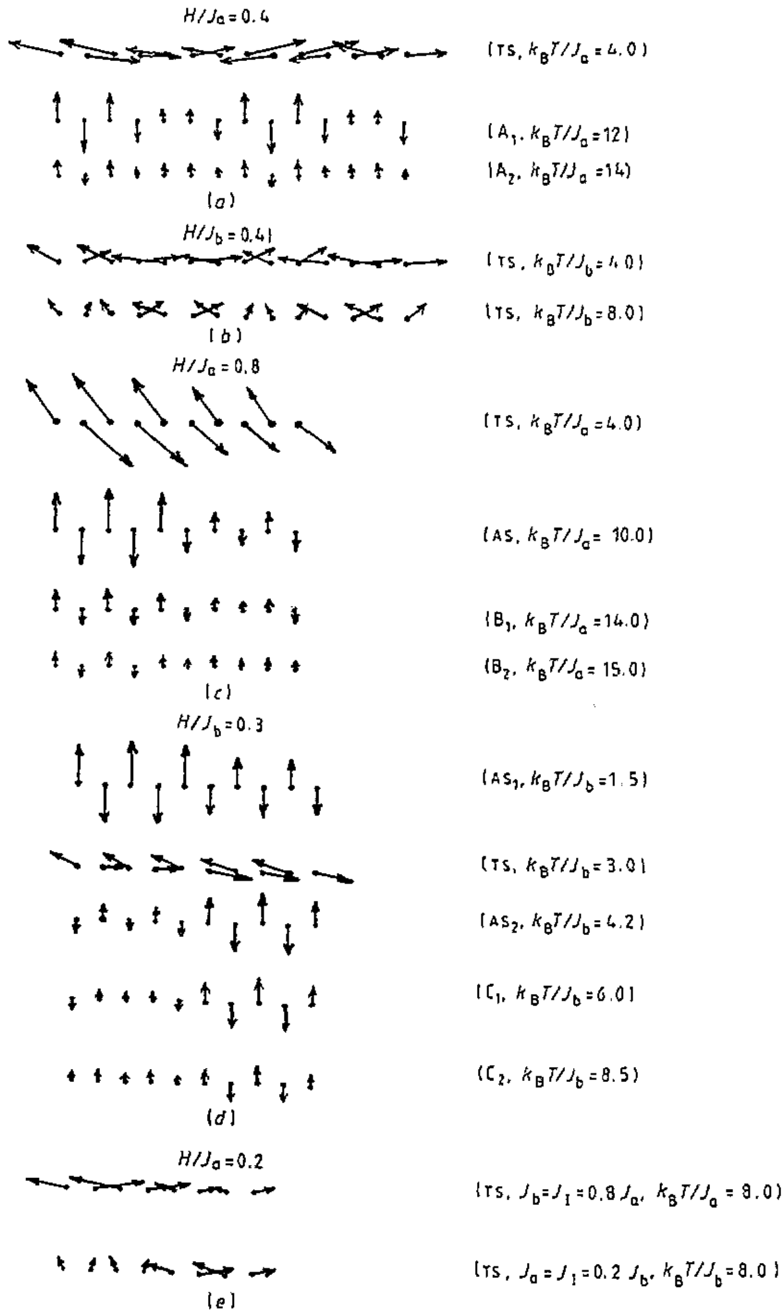

Figure 3. Spin configurations for the various phases of the different superlattices: (a) the 3_a+4_b superlattice with $J_b = J_I = 0.8J_a$; (b) the 3_a+4_b superlattice with $J_a = J_I = 0.2J_b$; (c) the 5_a+5_b superlattice with $J_b = J_I = 0.8J_a$; (d) the 5_a+5_b superlattice with $J_a = J_I = 0.2J_b$; (e) the 4_a+4_b superlattice.

of temperature. This, in part, shows the sharply rapid change in the twisted angles of atomic moments at the transition point. At the transition point from state B_1 to state B_2, the susceptibility curve has a small peak. In addition, we also see that the influence of the

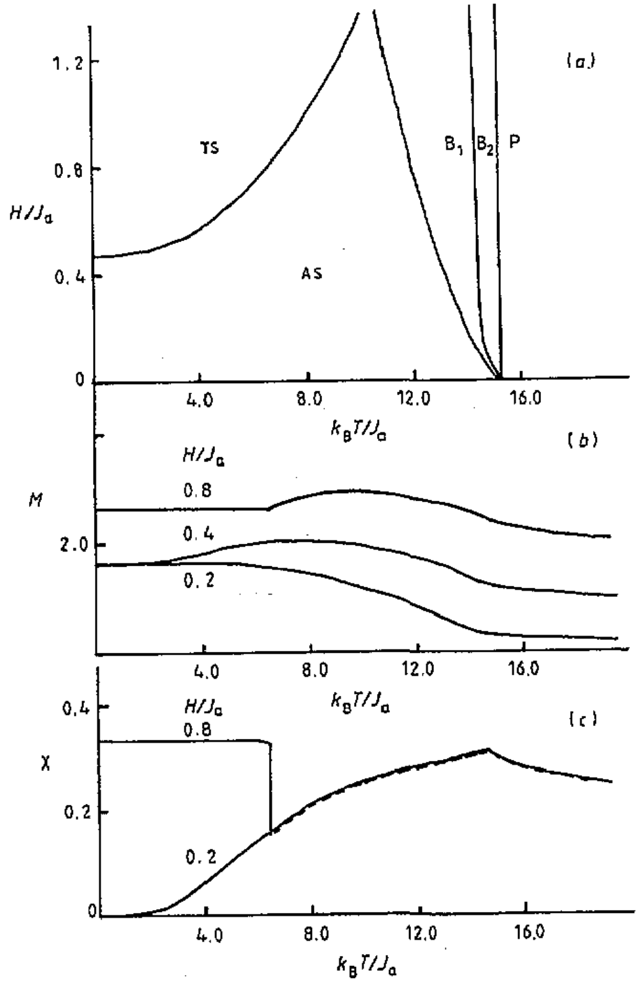

Figure 4. (*a*) The *H–T* phase diagram, (*b*) magnetization as a function of temperature, and (*c*) magnetic susceptibility as a function of temperature for the $5_a + 5_b$ superlattice with $J_b = J_I = 0.8J_a$. AS represents the antiparallel state.

external field on the susceptibility, except for the susceptibility in the twisted state, is slight.

It is more interesting that the $5_a + 5_b$ superlattice for case (2) can have six states: a twisted state, an antiparallel state 1 (AS_1), an antiparallel state 2 (AS_2), the states labelled C_1 and C_2, and a paramagnetic state. The spin arrangements related to these states are presented in figure 3(*d*). Although AS_1 and AS_2 are both antiparallel states, the directions of the spins in AS_1 are completely opposite to those in AS_2. At the same time, the difference between C_1 and C_2 is briefly that, in state C_2, all spins in the atomic slabs a are parallel to the direction of the external field but, in state C_1, the spins on the surfaces of the atomic slabs a are antiparallel to the direction of the field. Comparing figure 3 with figure 4, one can see the main differences between case (1) and case (2):

(a) There are two different antiparallel states.

(b) The magnetizations and magnetic susceptibilities in various states are more sensitive

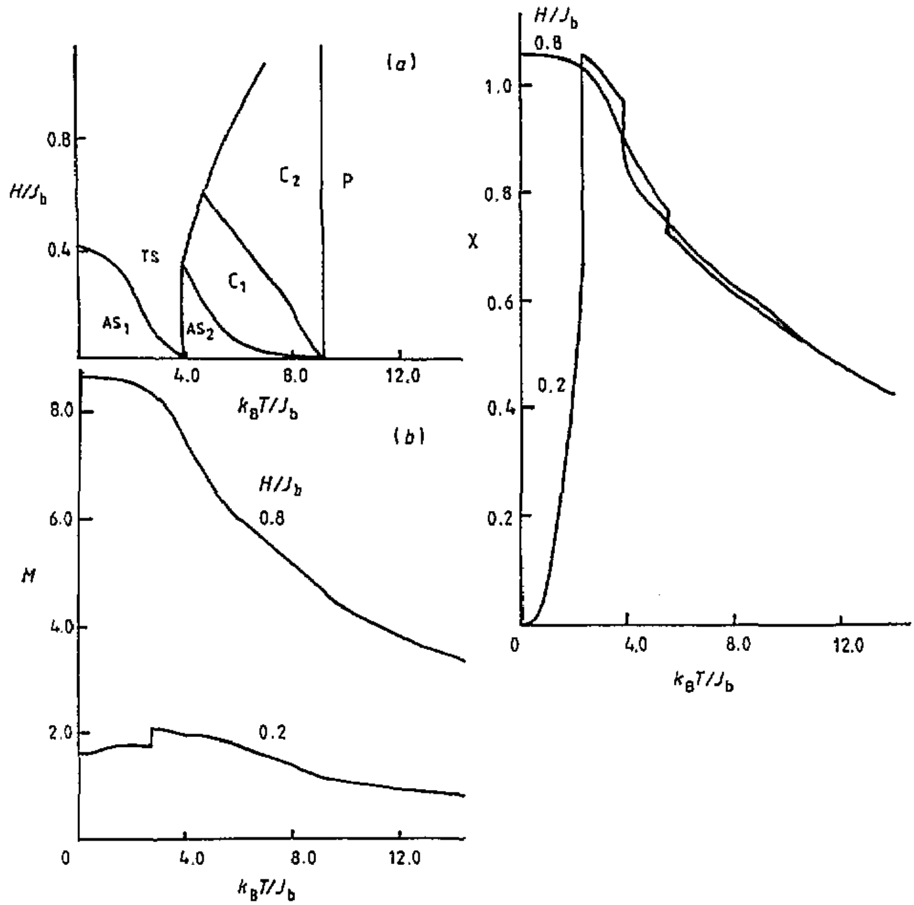

Figure 5. Properties of the $5_a + 5_b$ superlattice with $J_a = J_I = 0.2J_b$. (*a*)–(*c*) are as in figure 1. In (*a*) there are six states which are the antiparallel state 1 (AS_1), the antiparallel state 2 (AS_2), the twisted state (TS), states labelled C_1 and C_2, and the paramagnetic state (P). Note that in (*b*) the magnetization curve for $H = 0.2J_b$ is discontinuous at the transition point from AS_1 to TS.

to temperature.

(c) The magnetization as a function of temperature for the chosen external field $H = 0.2J_b$ is not continuous at the phase transition point from AS_I to the twisted state, which suggests that the transition is of first-order nature.

For the $3_a + 4_b$ and $5_a + 5_b$ superlattices, the common features are as follows.

(i) The macroscopic magnetic properties, the magnetization or magnetic susceptibility as a function of temperature, can reflect clearly the transition from a twisted state to another state (not a paramagnetic state).

(ii) The transition from some state to a paramagnetic state cannot be determined from these macroscopic magnetic properties, and this is practically important for determination of the 'Néel temperature' of these superlattices.

The phase diagrams for the $4_a + 4_b$ superlattice for different parameters J_a, J_b and J_I are qualitatively the same as the phase diagram in figure 2. If the external field is not very large, the transition point from the twisted state to the paramagnetic state is influenced only sightly by the field or this transition point is near the Néel temperature ($H = 0$) of the superlattice. Therefore, we do not present the phase diagrams in figure 6. Figures 6(a) and 6(b) show the magnetic susceptibility and magnetization, respectively, as functions of temperature. For $J_b = J_I = 0.8J_a$, the susceptibility depends on the external field in a similar way so that we only give one susceptibility curve for $h = 0.2J_a$ which is represented by the broken curve in figure 6(a). For $J_a = J_I = 2J_b$ the macroscopic magnetic properties are similar to those of the $3_a + 4_b$ superlattice with $J_a = J_I = 0.2J_b$.

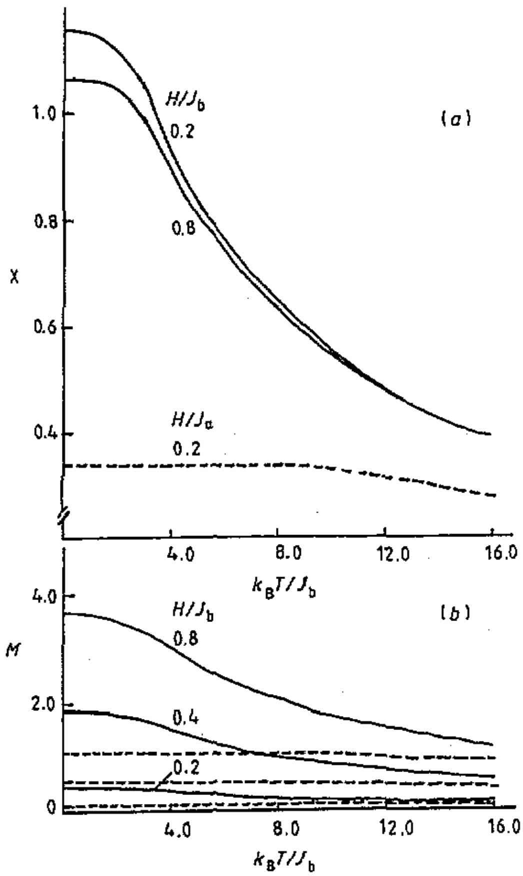

Figure 6. Magnetic properties of the $4_a + 4_b$ superlattice: (a) the magnetic susceptibility as a function of temperature; (b) the magnetization versus temperature. The broken curves correspond to $J_b = J_I = 0.8J_a$, and the full curves to $J_a = J_I = 0.2J_b$. Note that the curves are continuous at any temperature.

4. Summary

We have investigated the magnetization, magnetic susceptibility and phase diagrams of three typical superlattices formed from two completely different antiferromagnetic materials. Depending on the applied external fields, a variety of phases can appear in the $3_a + 4_b$ superlattice, composed of slab a with three atomic layers and slab b with four atomic layers, and the 5_a+5_b superlattice. The magnetic susceptibility (and also the magnetization in some cases) as a function of temperature can reflect clearly the transition from a twisted state to another state (except a paramagnetic state) but cannot show obviously the transition from any state to a paramagnetic state. This may be useful for experiments in which one estimates the Néel temperature of an antiferromagnetic superlattice. A special and interesting case is that, for the $5_a + 5_b$ superlattice with $J_a = J_I = 0.2J_b$, the magnetization as a function of temperature cannot be continuous at the transition point from AS_I to the twisted state. The isotropic Heisenberg model and the theory used in this paper are suitable for antiferromagnetic superlattices with a small anisotropy. For superlattices with a larger anisotropy, the results should be corrected, but we also see that our results for the magnetization (see figure 4(*b*)) of the $5_a + 5_b$ superlattice are similar qualitatively to some results for the FeF_2/CoF_2 superlattice with a 7/3 structure and a larger anisotropy in figure 2 of [11].

Acknowledgment

This work was supported financially by the Science Foundation of Heilongjing Province.

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