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Concentration dependence of local oscillations in Cu-Be system

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Inelastic scattering of cold neutrons was used to investigate the change of the phonon spectra of solid solutions of Be in Cu in the impurity concentration interval from 0.5 to 10 at.% with an aim at studying the transition from the local oscillations of a light isolated substitutional impurity to the impurity band. The measurements were made at room temperature. The experimental neutron-scattering cross sections were reduced in the noncoherent approximation. It was established that with increasing impurity concentration the following quantities vary linearly with the concentration: the energy position of the level of the local impurity oscillations, the width of this level, and the end-point frequency of the perturbed oscillation spectrum of the copper matrix.

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INTRODUCTION

Local oscillations (LO) produced in the phonon spectrum following introduction of a light impurity atom, or of an atom more tightly bound to the atoms of the host crystal (matrix), have been under investigation for a long time. They were predicted theoretically back in 1942 in the studies of I. M. Lifshitz.[1] LO were first observed experimentally in ionic crystals by optical methods, and in metallic systems with the aid of the Mössbauer effect and of inelastic neutron scattering.[2] Naturally, the observed distributions of the spectral density of the LO are not the δ functions predicted in the harmonic approximation for an isolated impurity. Nor is the LO lifetime, which is governed by the anharmonicity, the only factor that determines their real shape. Since any investigated system contains a finite impurity concentration, a definite role is played by the concentration broadening due to the interaction of the impurity atoms with one another.

However, there are practically no experimental investigations devoted to the concentration dependence of the LO width in metals where, unlike in ionic crystals, the restructuring of the electronic subsystem by introduction of an impurity influences substantially the character of the interaction between the atoms.

From among all the experimental methods of investigating impurity oscillations, inelastic scattering of slow neutrons is the most direct and most informative. The recently uncovered possibility of studying LO in metallic systems by tunnel experiments presupposes prior knowledge of the electron-phonon interaction function $\alpha^2(\omega)$, and is consequently not a direct method.

The purpose of the present study was to trace, with the aid of inelastic scattering of cold neutrons, the onset of LO and their variation in a metallic system, using as an example Cu-Be alloys at concentrations that range from a practically isolated substitutional impurity to concentrations such that the interaction of the impurity centers of the perturbation cannot be neglected, and an LO band appears. Of definite interest here is the change of the phonon spectrum of the perturbed matrix.

SAMPLES, MEASUREMENTS, AND DATA REDUCTION

The investigated Cu-Be system was chosen for a number of considerations. The dynamics of the copper lattice (matrix) has been sufficiently well investigated, as were also a number of physical properties of copper–beryllium alloys. Beryllium is an impurity with a single isotope. In addition, the LO due to the introduction of Be is as a rule far from the end point of the phonon spectrum of the matrix.[3-5]

The samples were made of specially purified copper containing not more than 10^{-3}% impurity. Binary copper alloys containing 0.5, 1, 2.7, 4.2, and 9.5 at.% Be, melted in vacuum, were homogenized at 1080 K for 10 hours and abruptly quenched to room temperature. The rolled sheets 1.5 mm thick were annealed in vacu-

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um at 640 K for 10 hours. A whole set of investigations (chemical analysis of the uniformity of the impurity distribution, x-ray diffraction, neutron diffraction, metallography, and small-angle scattering of cold neutrons) has shown that the prepared samples are practically homogeneous disordered solid solutions. Deviations of the impurity concentration at different points of the samples amounted to less than 3% than the average over the sample.

To assess the influence of the phase composition on the characteristics of the LO, additional samples were made, containing 18 and 27 at.% Be, more than the limiting concentration of the Be atoms in the α solution. In these two-phase samples, part of the impurity (9 and 23.% Be) turns out to be included in the CuBe intermetallide.

Measurements of the doubly differential cross sec tions for neutron scattering by Cu-Be alloys in pure copper were made with a time-of-flight spectrometer with a source of cold neutrons.[6] The energy of the neutrons incident on the sample was $E_0 = 4.8$ meV. The total energy resolution of the spectrometer in the LO region was 5.2% of the energy of the scattered neutrons. The measurements were made at room temperature simultaneously at six scattering angles Ω.

Figure 1 shows by way of example the energy spectra

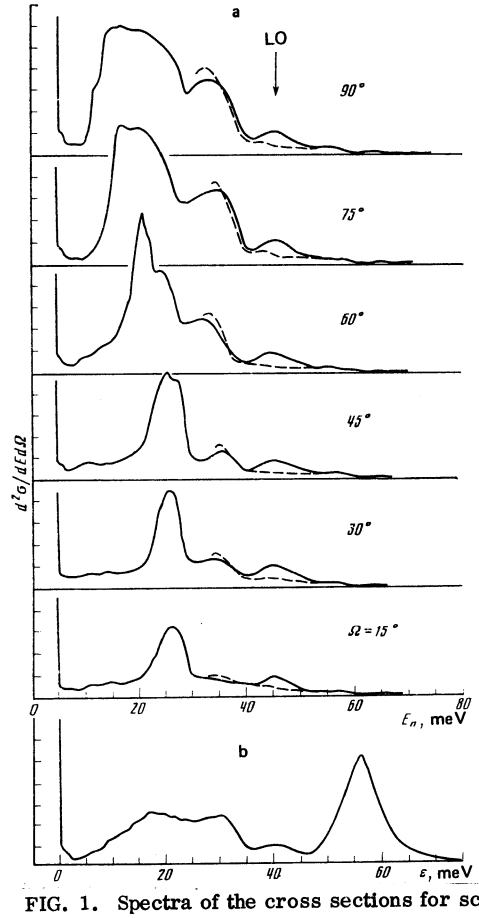

FIG. 1. Spectra of the cross sections for scattering of cold neutrons by the investigated Cu-Be alloys: a) α-solution, $c = 0.042$, the dashed line shows the part of the spectrum of scattering by pure copper; b) α + intermetallide CuBe, $c = 0.27$, angle $\Omega = 90°$.

of neutrons scattered by a disordered α-solution of $Cu_{0.958}Be_{0.042}$ and, for comparison, the spectrum of neutron scattered by a two-phase sample with a close Be concentration in the α solution. At an energy transfer $\varepsilon = E_n - E_0 = 40$ meV (E_n is the energy of the scattered neutrons), the spectrum for the α solution reveals distinctly a singularity due to the LO and missing from the spectrum of the Cu matrix. In the cross section for neutron scattering by the two-phase sample there appears an additional singularity at $\varepsilon = 57$ meV, due to optical oscillations in the spectrum for the intermetallide CuBe. This singularity is intense enough to serve as a criterion for the presence of the CuBe intermetallide phase in the investigated samples.[1)]

The reduction of the experimental LO spectra was carried out in the noncoherent approximation. The use of polycrystalline samples with disordered substitutional impurity atoms averages out the possible coherent effects completely. Taking into account the usual dependence of the cross section on the momentum transfer $\varkappa$, the generalized functions

$$\Theta(\varepsilon) = \frac{d^2\sigma}{d\varepsilon d\Omega}\varkappa^2 \exp[2W(\varkappa)], \tag{1}$$

measured at different scattering angles, agree with one another in the energy region of the LO within the limits of the statistical error of the experimental data.

Figure 2 shows the difference spectra $\Theta_{LO}(\varepsilon) = \Theta(\varepsilon)_{Cu-Be} - \Theta(\varepsilon)_{Cu}$, averaged over all the measurement angles. The normalization of the spectra that enter in the difference $\Theta_{LO}(\varepsilon)$, which is a function of the density of the squares of the displacements, was carried out over the noncoherent part of the inelastic neutron-scattering spectra in the energy region below the threshold of the Bragg scattering.

The possibility of using the noncoherent approximation for the experimental inelastic neutron-scattering cross section makes it possible to determine, without resorting to model representations, the integral characteristics of the spectral densities of the LO. The values of the LO energies (Fig. 3) and their error were calculated by the universally employed method.[7] Figure 3 shows also the end-point energies ε_{max} of the oscillations of the deformed lattice of the matrix atoms.

In the investigated concentration range, there are observed displacements of the end-point energy ε_{max} and of the position of the center of the LO spectral density (ε_{LO}) towards higher energies. The intensity (Fig. 4) and the experimental width (at half-height) of the LO spectrum (Fig. 5) increases linearly with increasing concentration of the beryllium atoms.

To take into account the distribution function, we used a method[8] that employs, as additional information, the assumption that the errors of the experimental data are statistically distributed. The most probable LO density functions are shown in Fig, 2 in the form of histograms. The relatively large error in the values of the true widths (Fig. 5) is due to the fact that the distributions of the LO spectral density were determined without using model representations concerning the functional form of these distributions.

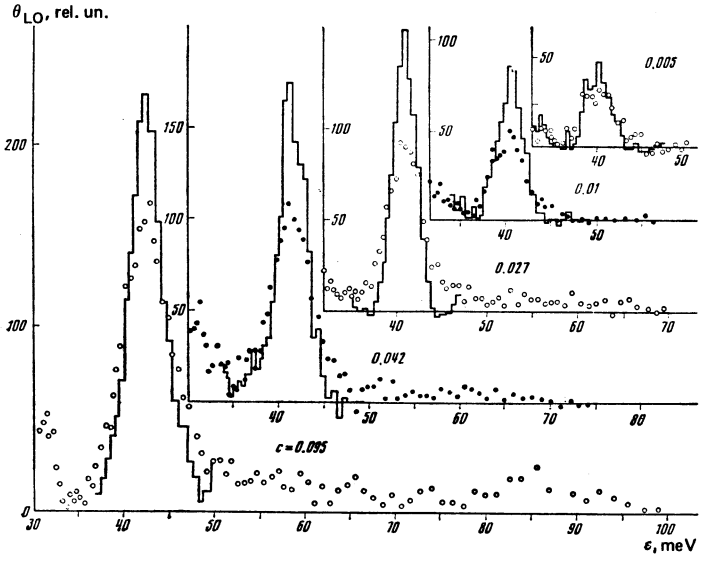

FIG. 2. Distribution function $\Theta_{LO}(\varepsilon)$ for different Be concentrations; points—experiment, histograms—reconstructed spectra.

When use is made of model calculations based on the assumption that the sought distribution has a Lorentz or Gaussian shape, a linear concentration dependence of the width of the LO is observed in the entire interval of investigated impurity-atom concentrations. The spectrometer resolution function, determined experimentally at energies close to ω_{LO} is well described by a Gaussian function. Therefore in the estimate of the widths of the model spectra we used the calculation formulas of Ref. 10. At a beryllium impurity concentration $c \geq 0.02$, satisfactory agreement between the widths reconstructed from the experimental data and the model calculations is obtained for a Lorentz curve.

DISCUSSION OF RESULTS

As seen from Figs. 3–5, the end-point energy of the vibrational spectrum of the deformed lattice matrix, the energy position, and also the integrated density of the LO states vary linearly with the concentration of the introduced beryllium atoms.

Extrapolation of the concentration dependence $\varepsilon_{LO} = f(c)$ to $c = 0$ yields the value of the LO energy for a strictly isolated impurity. This value turned out to be $\varepsilon_{LO,0} = 40.0 \pm 0.3$ meV, thus refining the result of Ref. 5, which was obtained with samples having a finite impurity concentration. The energy position of the LO differs substantially from the one calculated in the isotopic approximation (56 meV), thus testifying to a weakening of the effective constant of the impurity–matrix interatomic interaction, by an amount 49% compared with the initial interaction between the matrix atoms.

The displacement of the center of the LO spectral band ε_{LO} as a function of the concentration of the beryllium atoms is described by the relation

$$\delta\varepsilon_{LO}(c)/\varepsilon_{LO,0} = (0.68 \pm 0.06)c. \quad (2)$$

The concentration dependence of the change of the end-point energy of the matrix oscillation spectrum can be described by the relation

$$\overline{\delta\varepsilon_{max}(c)/\varepsilon_{max}(0)} = (0.51 \pm 0.05)c. \quad (3)$$

An estimate of the displacement of the end-point fre-

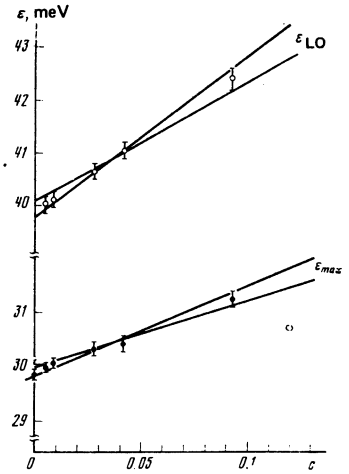

FIG. 3. Concentration dependence of the LO energies and of the end-point of the principal spectrum. The lines determine the limiting values of the coefficients in relations (2) and (3).

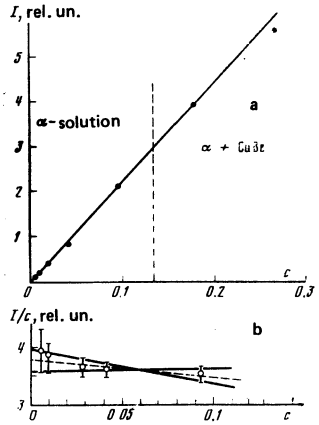

FIG. 4. Concentration dependence of the intensity of neutron scattering by oscillations due to the beryllium atoms. The dashed line in Fig. 4b corresponds to $f(c) = (1 - c)f(0)$.

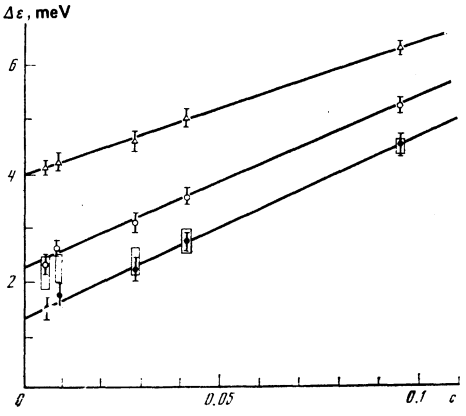

FIG. 5. Concentration dependence of the widths of the spectral distributions: Δ—experiment, ○—Gaussian, ●—Lorentzian, rectangles—model-independent reconstruction. The heights of the rectangles correspond to the errors in the calculation results.

quency because of the change in the volume of the unit cell, under the condition that the Grüneisen constant γ for copper and its alloys is equal to 2, yields a concentration dependence

$$\delta\omega/\omega=\gamma\Delta V/V=0.53c. \quad (4)$$

Thus, the displacement of the end-point frequency of the continuous spectrum of the oscillations is due to the practically complete effect of compression of the lattice, which is accompanied by the introduction of the beryllium atoms.

We consider now the concentration dependence of the ratio

$$\frac{\varepsilon_{LO}(c)}{\varepsilon_{max}(c)}\approx\frac{\varepsilon_{LO,0}}{\varepsilon_{max}(0)}\left[1+\frac{\delta\varepsilon_{LO}(c)}{\varepsilon_{LO,0}}-\frac{\delta\varepsilon_{max}(c)}{\varepsilon_{max}(0)}\right], \quad (5)$$

which describes the relative change of the interatomic force interaction in the alloy. The weak change of this ratio with increasing beryllium concentration [the values (2) and (3) were substituted in (5)] indicates a negligible increase of the interaction constant β_{Be-Cu} compared with the averaged constant $\bar{\beta}$ of the alloy. Therefore, if the change $\Delta\beta/\bar{\beta}$ is attributed to a change of the screening charge of the impurity, which is determined by the density of the conduction electrons,[11] then a relatively weak change of the electron density over the alloy is observed.

In the case of two-phase samples, attention is called to the fact that the energy position of the level of the optical oscillations coincides with the expected value of ε_{LO} in the isptopic substitution model. The agreement between the values of ε_{LO} is apparently due to the fact that the principal assumption of the isotopic-substitution model, namely that the interatomic force interaction averages out,[12] is fully realized in electronic compounds of this type.

The ratio of the density of the LO states to the density of the oscillations of the matrix is proportional, in first-order approximation, to the concentration of the beryllium impurity. However, the intensity of the scattered neutrons, normalized correspondingly to the same number of impurity and matrix atoms, is larger by a factor of 3.6 for the LO. In addition, the measured cross section for neutron scattering by the LO is double the cross section calculated with allowance for the perturbation of the lattice in the isotopic approximation.[13] The difference is probably due to some delocalization of the oscillations of the impurity atoms and the additional involvement of about two matrix atoms by these oscillations. This quantity characterizes the degree of damping of the LO, in which there participate on the average 1/6 of the copper atoms located in the first coordination sphere in the fcc lattice of the Cu-Be α-solution. An estimate of the damping constant ($a=\ln 6(r_{Cu}+r_{Be})^{-1}\approx 2.7d^{-1}$, where d is the matrix lattice parameter) has shown, that the weakening of the interaction of the impurity atoms with the matrix in the Cu-Be system leads to the same degree of damping of the local perturbation as in Cu-Al alloys[14] which have lower component-atom masses and respectively lower ε_{LO} and ε_{max}.

With increasing impurity concentration, when the distance between the impurity centers decreases, the perturbed regions can overlap, i.e., the cross sections for the scattering by the perturbed regions become non-additive, and the concentration dependence of I/c (Fig. 4b) is close to the Nordheim formula: $f(c)=1-c$.

The character of the dependence of the intensity of the scattered neutrons on the Be concentration is preserved in the entire investigated interval of impurity-atom concentrations, including two-phase systems (Fig. 4a). The probable reason is that we have a weak energy dependence of the polarization vectors for the optical oscillations in the case of diatomic systems with so greatly differing masses as Be and Cu. This circumstance was noted earlier[15] for the behavior of hydrogen in a metal.

As already indicated, the widths of the reconstructed spectra (Fig. 5) for alloys with $c\geq 0.02$ are close to the widths obtained under the assumption that the sought spectral line has a Lorentz shape. This is evidence that the principal mechanisms in the formation of the LO spectrum are those leading to homogeneous broadening. Extrapolation to $c=0$ determines the lifetime of the LO of the localized impurity: $\tau=2.6\cdot 10^{-12}$ sec, a value limited by the decay of the LO into two phonons of the principal spectrum ($\varepsilon_{LO}<2\varepsilon_{max}$) and by the interaction of the LO with the electrons. Since the density of the electronic states on the Fermi surface is small for copper, and the electron-phonon interaction depends quadratically on the density of the electronic states, the width of the LO level for an isolated impurity determines completely the anharmonicity temperature constant of third order. The obtained damping $\Gamma=0.028\ k_BT$ is typical of high-frequency phonons in metal lattices.[16]

The question of formation of the vibrational spectrum of disordered systems, including the case when light impurity atoms are introduced, was considered theoretically in a large number of papers.[1,17-20] It turned out that the results of these studies are very sensitive to the character of the assumptions made, and frequently lead to contradictory conclusions. Thus, in the coherent-potential approximation the width of the LO band de-

pends on the impurity concentration like $c^{1/2}$, and in the approximation of the non-self-consistent t-matrix the dependence is linear, but the shape of the LO spectrum in this approximation is asymmetric and reflects the singularities of the spectrum of the host matrix.[17] In an analysis of the indirect dynamic interaction between impurity atoms, the authors of Ref. 19 conclude that the width of the LO spectrum has a very strong concentration dependence. In Ref. 20 it is stated flatly that analytic methods that employ Green's-function expansions are not valid for singular points of phonon spectra of an impurity crystal, particularly for local states. Identifying the width of the band in which the group expansion of the Green's function diverges with the width of the impurity band, the authors of Ref. 20 obtain for it a $c^{2/3}$ dependence, and an $\exp\{-\alpha c^{-1/3}\}$ as $c \to 0$.

Taking into account the concentration dependence observed in the width of the LO spectrum of the investigated Cu-Be system, it is preferable to compare the obtained data with results calculated in the t-matrix approximation. In this approximation, the total width P of the LO band, the expression for which was obtained without allowance for the change in the force constants[21]:

$$P = \omega_{LO} \frac{c}{2\xi(1-c)^2} \left[\frac{\omega_{LO}^2}{\omega_{max}^2} - 1 \right]^{-1} \approx 2\Delta\omega_{LO} \qquad (6)$$

where $\xi = 1 - m_{Be}/m_{Cu}$, has practically a linear concentration dependence. Using relation (6) in the experimental value of $\omega_{LO,0}$ we obtain for $\Delta\varepsilon_{LO}$ a concentration dependence which is approximately half that observed in Fig. 5.

The character of the change of the structure of the spectral density of the LO with the beryllium concentration can be traced most clearly by considering the normalized difference spectra (Fig. 6):

$$\Delta g(\varepsilon, c)/\Delta c = [\Theta_{LO}(\varepsilon, c_i) - \Theta_{LO}(\varepsilon, c_{i-1})](c_i - c_{i-1})^{-1}. \qquad (7)$$

At low concentrations the character of the distribution remains unchanged, but some shift is observed towards higher energies. With increasing concentration of the impurity atoms there occurs not only a displacement of the band, but also a substantial broadening on account of the predominant increase of the density of the high-energy states. The observed character of the change of the spectral density of the LO is evidence of the different mechanisms of the interaction of the impurity atoms with one another as functions of the concentrations of the latter. At low concentrations, i.e., when the average distance between the impurity atoms $\bar{r} \sim c^{-1/3}$ is much larger than the lattice constant, the principal mechanism is indirect dynamic interaction via the matrix atoms. With increasing impurity concentrations, the decisive mechanism is already the indirect static interaction due to the overlap of the perturbation regions; this corresponds to the classical broadening mechanism considered by Lifshitz.[1]

The presence of a second phase in the form of intermetallic inclusions, up to 9 at.% Be, does not lead to an additional broadening of the LO band (Fig. 7). It appears that the mutual influence of the two phases is limited to a narrow region near their separation boundary, and the relative contribution of this region is small.

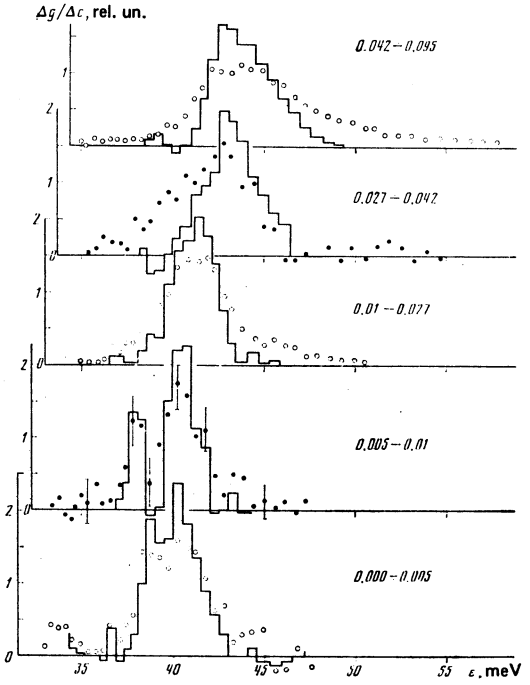

FIG. 6. LO difference spectra. Points—experiment, histograms—model-free reconstruction. On the right are marked the values of c_{i-1} and c_i.

Thus, we have investigated with the aid of inelastic neutron scattering, for the first time ever, the influence of the concentration mechanism on the formation of local states of metallic systems. We have shown that the principal characteristics of the LO spectrum (density, position, width) are linearly connected with the concentration of the impurity atoms. This concentration dependence of the principal characteristics of the LO cannot be satisfactorily explained within the framework of any of the existing theories. No splitting of the LO band similar to that observed in Ref. 3 for V-Be alloys occurs up to impurity-atom concentrations $c = 0.1$. At low concentrations, the lifetime of the LO is

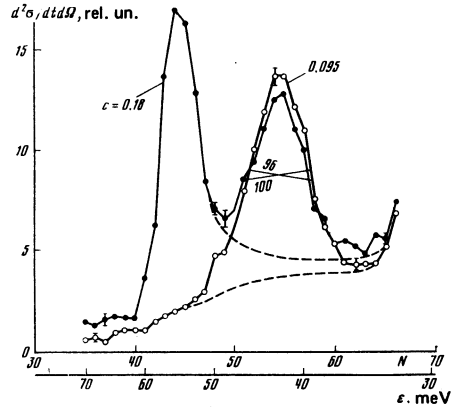

FIG. 7. Spectral distributions of the oscillations due to Be atoms in one-phase (○) and two-phase (●) Cu-Be samples. The widths of the LO at half-height are given in microseconds. N is the number of the time-analyzer channel.

determined by the anharmonicity. The anharmonicity constant for the LO turned out to be close to the value of the constant for the high-frequency oscillations of the crystal lattice.

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Nonlinear waves and the dynamics of domain walls in weak ferromagnets

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Nonlinear waves are investigated in a two-sublattice antiferromagnet with noncollinear sublattices. Simple magnetization waves, describing the motion of 180-degree domain walls of two types, are considered far from the spin-flip region. The specific nature of nonlinear magnetization waves within the spin-flip region is investigated. Solutions are obtained that describe two-parameter magnetic solitons, to which corresponds a periodic oscillation of the magnetization in a reference system moving with the wave. The properties of N-soliton solutions, describing the interaction of nonlinear waves, are discussed. Formulas are obtained that describe the velocity of steady-state motion of a domain wall under the action of an external magnetic field.

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In a description of the nonlinear dynamics of magnetically ordered crystals, there almost always arises the problem of the properties of isolated magnetization waves. The interest in this problem is due to the broad possibility of describing the nonlinear dynamics of a magnetic material in terms of nonlinear waves. In particular, nonlinear isolated waves describe an effect that is important practically: the motion of domain walls (DW) and of isolated magnetic domains during the magnetization reversal of magnets.[1]

The theory of nonlinear waves has been developed in greatest detail for the case of a magnet with a single sublattice. It is known, however, that in magnets with two equivalent sublattices the dynamics of nonlinear waves is different in a number of important features. Among the specific features of this system must be included the exchange character of the limiting velocity of motion of DW,[2,3] which attains tens of kilometers per second,[4-6] and the presence of certain types of nonlinear waves that correspond to single boundary conditions.[2,7]

A very interesting class of such magnets is the weak ferromagnets (WFM) (in particular, the rare-earth orthoferrites (REO)), in which the Dzyaloshinskiĭ interaction leads to noncollinearity of the sublattices; that is,

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