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V.I.Yukalov

MÖSSBAUER-EFFECT PROBABILITY FOR HETEROGENEOUS MATTERS

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When measuring the Mössbauer-effect probability near phase-transition points, a common for different matters phenomenon was observed - this probability showed a minimum at the transition point. For example, such a sagging in the effect probability as a function of the temperature occurs at the Curie point of magnets. The most detailed investigation of this effect was done in paper [1] for ferrite-garnets $Y_3Fe_5O_{12}$ and $Y_3Fe_4A\ell O_{12}$.

The diminishing of the Mössbauer-effect probability could be naturally connected with a diminishing of the effective Debye temperature T_D. However, it has not been clear, which microscopic reason this diminishing is caused by. One could try to explain the change of T_D by the corresponding changes in the phonon spectrum owing to the appearance of a magnetic order. But, as is shown by a number of independent experiments [2-7], the relative change of phonon characteristics below the Curie point T_C does not exceed 1% as compared with the paramagnetic region. Such a change is too insignificant for explaining the diminishing of f_M by about 10%. Moreover, the measurement accuracy of f_M is also not better than 1%, therefore the effects connected with the spin-phonon interaction are not observable with such an accuracy.

An analogous sagging of the Mössbauer effect probability was found [8] in the ferroelectric $BaTiO_3$. The attempts to explain this sagging by the existence of a soft mode in the phonon spectrum, as it turned out, were flimzy - Binder, Meissner and Mais [9,10] analysed in detail the situation and proved that the appearance of a soft mode led solely to a fracture of f_M as a function of the temperature but by no means to a sagging.

Similar saggings of f_M were observed as well [11,12] under reoriental tional transitions near the compensation point in ferrite garnets $Y_{1.2}Gd_{1.8}Fe_5O_{12}$ and $Gd_3Fe_5O_{12}$ and hexaferrites $Ba_{2-x}Sr_xZn_2Fe_{12}O_{22}$ (Sr_xZn_2Y).

The diminishing of the effect probability f_M was also measured after the irradiation of a matter by fast neutrons. Sinitsin, Spirin and Upensky [13], for instance, studied this effect for ferrite garnets $Y_3Fe_5O_{12}$ and $Y_3Fe_{5-x}A\ell_xO_{12}$.

In the present paper, the point of view is proclaimed and proved according to which all the phenomena mentioned above are caused by one common reason - the heterogeneity of the matter. This heterogeneity either is connected with the appearance of heterophase fluctuations near phase-transition points [14-18], or is directly due to the neutron irradiation of the matter.

Really, the fact that in the vicinity of a phase-transition point the matter is, generally speaking, a mixture of competing phases was repeatedly demonstrated by various methods, including the Mössbauer spectroscopy. The existence of a nonmagnetic phase in a magnet at temperatures $T < T_c$, or vice versa, of magnetic clusters in a paramagnet above T_c was clearly fixed by the form of the Mössbauer spectrum having side by side with the sixline magnetic component the singleline component corresponsing to the paramagnetic phase. The presence of heterophase states in magnets near the transition point magnet-paramagnet was experimentally checked by means of the Mössbauer spectroscopy for a number of matters: it was found in Fe and Ni [19-21], in the isotope of Fe with spin 3/2 [22], in the alloys $Zn_xNi_{1-x}Fe_2O_4$ [23] and $KFeS_2$ [24], and other materials.

The existence of clusters of competing phases near the transition point ferroelectric-paraelectric is also very well known for many ferroelectrics [25-28].

The presence of a mixture of several phase with different directions of magnetization in the vicinity of reorientational transitions is experimentally established [29,30] as well.

Finally, the irradiation of solids by fast neutrons leads, as is known, to the formation of the so-called regions of disorder having essentially a lower density than the surrounding matter. These regions of disorder are nothing but the nuclei of the phase that is called disordered or amorphised. The concentration of the new phase can be also measured with the help of the Mössbauer spectroscopy [13].

It is just this common property, i.e. the heterogeneity, that is, from our point of view, the main reason for the diminishing of the Mössbauer effect probability in all situations discussed above. The confirmation of this point of view is given below. The system of units $\hbar = 1 = k_B$ is used.

When the matter investigated consists of two phase, the Mössbauer effect probability is of the form

$$f_M = w_1 f_1 + w_2 f_2 , \tag{1}$$

in which w_α is the concentration of the phase α, w_α is defined by minimizing a thermodynamic potential [14-18], f_α is the effect probability for the corresponding phase,

$$f_\alpha = \exp(-2W_\alpha) , \tag{2}$$

the Lamb-Mössbauer factor being

$$W_\alpha = \frac{3k^2T^2}{mT_\alpha^3} \int_0^{T_\alpha/2T} \omega \,\mathrm{cth}\,\omega \, d\omega , \tag{3}$$

here k is the gamma-quantum wave vector, m is the mass of a Mössbauer nucleus, T_α is the effective Debye temperature for the considered matter in the phase α in the case of a mixture of several phases, T_α is connected with the Debye temperature T_D of the pure phase by the relation [16]

$$T_\alpha = w_\alpha^{1/2} T_D . \tag{4}$$

In what follows the index $\alpha = 1$ corresponds to the ordered phase and $\alpha = 2$ to the disordered one. We also use the notation

$$w_2 = x , \quad w_1 = 1 - x , \tag{5}$$

taking account of the normalization condition

$$w_1 + w_2 = 1 .$$

Formulae (2)-(5) shows that the effect probability (1) is a function of the concentration x,

$$f_M = f_M(x) .$$

The relative change of the effect probability for the heterophase matter as compared with the pure one is characterized by the quantity

$$\delta f_M(x) = \frac{f_M(x) - f_M(0)}{f_M(0)} . \tag{6}$$

Consider first the transition paramagnet magnet. When approaching the critical point from above in the paramagnet, beginning from the upper nucleation point T_n, the clusters of magnetization appear [21]. At the critical point itself both the phases are indistinguishable, so $x = 1/2$. The nuclei of the paramagnetic phase survive below the critical temperature [19-24] down to the lower nucleation point T_n. The width of the heterophase region observed in experiments

$$\delta T \equiv (T_n' - T_n)/T_c ,$$

varies from one matter to another. For instance, in Fe and Ni this width is of the order $\delta T \sim 10^{-3}$ (see [19,20]), in the 3/2-spin isotope Fe [22] and in the alloys $Zn_x Ni_{1-x} Fe_2 O_4$ [23] the coexistence region of the antiferromagnetic and paramagnetic phases is characterized by $\delta T \sim 10^{-1}$, in the antiferromagnetic alloy $KFeS_2$ the paramagnetic phase is present at all temperatures $T > 0$ disappearing only at $T = 0$ [24]. Therefore, the concentration of the paramagnetic phase at these characteristic points is

$$x(T_n) = 0, \quad x(T_c) = \frac{1}{2}, \quad x(T_n') = 1.$$

The variation of the effect probability (6) for the critical point T_c, where $x = 1/2$, can be found using eqs.(1)-(4), hence,

$$\delta f(\tfrac{1}{2}) \simeq \begin{cases} f_M^{0.4}(0) - 1, & T_c \ll T_D, \\ f_M(0) - 1, & T_c \gg T_D. \end{cases} \qquad (7)$$

Compare (7) with data on ferrite-garnets [1] with the relative width of the heterophase region $\delta T \sim 10^{-3}$ and $f_M(0) \simeq 0.7$. Then (7) gives

$$\delta f(\tfrac{1}{2}) \simeq \begin{cases} -0.1, & T_c \ll T_D, \\ -0.3, & T_c \gg T_D. \end{cases} \qquad (8)$$

For the case $T_c \gtrsim 2\pi T_D$ corresponding to the ferrite garnets considered [1] $\delta f(1/2)$ given by (6) is $\delta f(1/2) \simeq 0.3$. Such a sagging of the Mössbauer effect probability is in good agreement with the experiment [1]. The saggings of the effect probability at the critical point of ferroelectrics [8] and at the compensation point [11,12] are almost the same.

Calculate now the diminishing of the Mössbauer effect probability after irradiating solids by fast neutrons. The usual concentration of the disordered phase is quite small, $x \ll 1$. Then (6) yields

$$\delta f(x) \simeq -x \begin{cases} 1 + W, & T \ll T_D, \\ 1 + 2W, & T \gg T_D, \end{cases} \qquad (9)$$

where W is the Lamb-Mössbauer factor for a pure phase,

$$W = \frac{3k^2 T^2}{m T_D^3} \int_0^{T_D/2T} \omega \coth\omega \, d\omega = \begin{cases} 3k^2/8mT_D, & T \ll T_D, \\ 3k^3\, T/2mT_D^2, & T \gg T_D. \end{cases}$$

As far as practically always

$$k^2/mT_D \ll 1,$$

then $W \ll 1$, therefore in place of (9) we have for all temperatures

$$\delta f(x) \simeq -x \qquad (x \ll 1). \qquad (10)$$

In the experiments on ferrite-garnets [13] the dependence of quantity (6) on the concentration of the disordered (amorphised) phase has been analyzed at temperatures from 80 up to 670K; the value of the concentration being varied in the interval $x \simeq 0.05 - 0.1$ owing to variations of irradiation doses. The experimental data [13] within the accuracy of measurements (4%) coincide with the theoretical relation (10).

Such a good agreement with experiment confirms that in all the considered cases the diminishing of the Mössbauer-effect probability can be explained as due to one common reason - the heterogeneity of the matters.

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Юкалов В.И. E17-88-582

Вероятность эффекта Мёссбауэра
для гетерогенных веществ

Исследовано совместное влияние гетерофазных флуктуаций и фононных возбуждений на решеточные и магнитные свойства ферромагнетиков. Для введения фононных переменных использована процедура, сохраняющая трансляционную симметрию. Спиновые переменные рассматриваются в приближении Тер-Хаара и в приближении среднего поля. Находится перенормировка скорости звука, обусловленная как спин-фононной связью, так и гетерофазными флуктуациями; численные оценки показывают, что последние играют решающую роль вблизи критической точки. Вычислена вероятность эффекта Мёссбауэра и предложено объяснение аномального прогиба в точке перехода.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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Yukalov V.I. E17-88-582

Mössbauer Effect Probability
for Heterogeneous Matters

An analysis is given of the following two questions; the sagging of the Mössbauer effect probability which has been observed in different matters at phase-transition points such as magnet paramagnet, ferroelectric paraelectric and reorientational magnetic transitions; the diminishing of this probability resulting from irradiation of solid. It is shown that the diminishing of the Mössbauer effect probability in all these cases can be explained by a common reason the heterogeneity of the matter. Calculations made for the mixture magnet paramagnet and for a solid with regions of disorder show a nice agreement with experiment.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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