IMPURITY SPIN-WAVE STATES IN NONCOLLINEAR ANTIFERROMAGNETS

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We consider the dynamics of a Heisenberg antiferromagnet containing an isolated point defect, in which the noncollinearity of the magnetic moments of the sublattices is produced by an external magnetic field. We determine the dependence of the resultant impurity oscillations on the applied field H. We show that quasilocal oscillations of the s type are revealed experimentally by the splitting of the AFMR line.

IN recent experiments performed by Borovik-Romanov and Meshcheryakov,[1] a splitting of the antiferromagnetic resonance (AFMR) lines was observed in $CoCO_3$ at a frequency on the order of 46 GHz in a magnetic field of approximately 3 kOe. An investigation of the chemical composition of the samples in which this effect was observed has shown that they contain iron as an impurity, and the magnitude of the splitting increases with increasing concentration of the iron atoms, while the resonant frequency is practically independent of the concentration. The temperature dependence of the resonant frequency is the same as the analogous dependence of the sublattice magnetization. This might suggest that quasilocal states of spin waves were observed in these experiments.

The question of the possible existence of impurity magnetic states in ferromagnets and antiferromagnets with antiparallel orientation of the magnetic-sublattice moments was considered theoretically in [2,3], where the model of local perturbation, first proposed by I. Lifshitz,[4-8] was employed. It is precisely the assumption that the interaction is local, and not that it is small, which is of importance in the investigation of the energy spectrum of nonideal crystals.

In the present paper we consider the question of the existence of local and quasilocal magnetic oscillations in a noncollinear Heisenberg antiferromagnet containing a magnetic impurity of the substitutional type and having the symmetry of a body-centered cube. We find the energy spectrum of such a crystal in a wide range of magnetic fields for ferromagnetic and antiferromagnetic impurities. We show that the quasilocal oscillations of the s type can interact in resonant fashion with the homogeneous-precession spin wave, and can therefore be revealed experimentally by the splitting of the AFMR line.

1. HAMILTONIAN OF NONIDEAL CRYSTAL

We consider a Heisenberg antiferromagnet having the structure of a body-centered cube (Fig. 1) and containing an isolated magnetic substitutional impurity. We consider, in addition, only the exchange and Zeeman energies. Such a model can describe sufficiently well antiferromagnets with anisotropy of the "easy-plane" type ($MnCO_3$, NiF_2, $CoCO_3$), and also antiferromagnets with anisotropy of the "easy-axis" type (α-Fe_2O_3, Cr_2O_3), in magnetic fields exceeding the critical field of the "turning over" of the magnetic moments of the sublattices. If we confine ourselves only to the interaction between

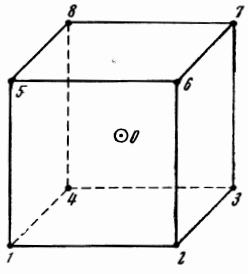

FIG. 1. Arrangement of the atoms closest to the impurity atom.

nearest neighbors, then the Hamiltonian of the system in question takes the form

$$\mathcal{H} = J\sum_{n\Delta} S_{1n+\Delta}S_{2n} - \mu gH\sum_{n}(S_{1n}^{z} + S_{2n}^{z}) + \sum_{\Delta}(J'S_{1\Delta}S_{20}' - JS_{1\Delta}S_{20}) - \mu H(g'S_{20}'^{z} - gS_{20}^{z}),$$

where J and J′ are the values of the exchange integrals of the matrix and impurity atoms; g, g′ and S, S′ are the corresponding g factors and spins of the ideal and impurity atoms; H is the external magnetic field and μ the Bohr magneton; the vector **n** runs through all the lattice sites, **Δ** is a vector designating the atom closest to the given one; 1 and 2 are the indices of the magnetic sublattices.

In a magnetic field **H**, the sublattice magnetic moments become noncollinear, and it is therefore more convenient to change from the coordinate system (x, y, z) with the z axis directed along the field **H**, to a new system (ξ, η, ζ), by means of the relations

$$S_n^{x} = S_n^{\xi};\quad S_n^{\eta} = S_n^{\eta}\cos\theta_n + S_n^{\xi}\sin\theta_n;\quad S_n^{z} = -S_n^{\eta}\sin\theta_n + S_n^{\zeta}\cos\theta_n. \tag{2}$$

Here θ_n is the angle between the direction of the equilibrium position of the spin at the site **n** and the field **H**.

To find the weakly-excited states of the antiferromagnet, we use the Holstein-Primakoff transformation[9]

$$S_n^{\xi} = \sqrt{\frac{S_n}{2}}(a_n + a_n^{+}),\quad S_n^{\eta} = -i\sqrt{\frac{S_n}{2}}(a_n - a_n^{+}),\quad S_n^{\zeta} = S_n - a_n^{+}a_n, \tag{3}$$

after which the Hamiltonian (1) takes the form

$$\hat{\mathcal{H}} = E + \hat{\mathcal{H}}_0 + V, \tag{4}$$

where the energy of the ground state is

$$E = JS^2\sum_{n\Delta}{}'\cos(\theta_{1n+\Delta} - \theta_{2n}) - \mu gHS\sum_{n}{}'(\cos\theta_{1n} + \cos\theta_{2n}) + J'S'S\sum_{\Delta}\cos(\theta_{1\Delta} - \theta_{20}) - \mu g'HS'\cos\theta_{20}, \tag{5}$$

and $\hat{\mathcal{H}}_0$ and $\hat{V}$ are respectively the Hamiltonian of the ideal crystal and the operator of the perturbation introduced by the impurity; Σ' denotes summation over all lattice sites with the exception of the impurity site. The equilibrium values of the angles θ_n can be determined in principle by minimizing (5) with respect to θ_n. However, the solution of the system of equations obtained in this case is very difficult, and we confine ourselves to an approximation in which the θ_n differ from the corresponding angles of an ideal crystal only at the impurity site and on the first coordination sphere. Such an approximation should be sufficiently good, since the deviation of θ_n from the angle in the ideal crystal decreases with increasing distance to the impurity like z^{-k}, where z is the number of atoms closest to the given one and k is the number of the coordination sphere in which the n-th atom is situated. In this case the operators $\hat{\mathcal{H}}_0$ and $\hat{V}$ take the form

$$\hat{\mathcal{H}}_0 = JS\sum_{n\Delta}[a_{1n}^+a_{1n} + a_{2n}^+a_{2n} + \cos^2\theta\,(a_{1n+\Delta}^+a_{2n} + a_{2n}^+a_{1n+\Delta}) + \sin^2\theta\,(a_{1n+\Delta}a_{2n} + a_{2n}^+a_{1n+\Delta}^+)], \tag{6}$$

$$\hat{V} = \sum_{\Delta}[v_1a_{20}^+a_{20} + v_2a_{1\Delta}^+a_{1\Delta} + v_3(a_{1\Delta}^+a_{20} + a_{20}^+a_{1\Delta}) + v_4(a_{1\Delta}a_{20} + a_{20}^+a_{1\Delta}^+)], \tag{7}$$

where the values of v_i are given by

$$\begin{aligned} v_1 &= {}^1/_8\,\mu H(g'\cos\theta_{20} - g\cos\theta) + [J\cos 2\theta - J'\cos(\theta_{1\Delta} - \theta_{20})]S,\\ v_2 &= \mu gH(\cos\theta_{1\Delta} - \cos\theta) + JS\cos 2\theta - J'S'\cos(\theta_{1\Delta} - \theta_{20}),\\ v_3 &= {}^1/_2J'\sqrt{SS'}\,[1 + \cos(\theta_{1\Delta} - \theta_{20})] - JS\cos^2\theta,\\ v_4 &= {}^1/_2J'\sqrt{SS'}\,[1 - \cos(\theta_{1\Delta} - \theta_{20})] - JS\sin^2\theta. \end{aligned} \tag{8}$$

In formulas (8), θ is the angle between the equilibrium position of the spin of the first sublattice and the direction of the magnetic field in the ideal crystal, and is determined by the well-known relation

$$\cos\theta = \mu gH/16JS. \tag{9}$$

The dependence of the angles θ_{20} and $\theta_{1\Delta}$ on the field H is shown in Fig. 2 for the following values of the impurity parameters: J'/J = −0.5, 0, 0.5, and g'/g = 0.606.

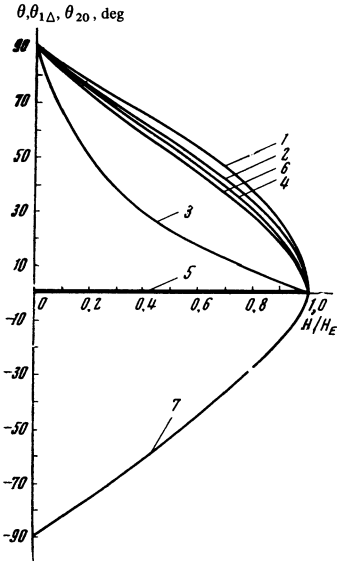

FIG. 2. Dependence of the angles θ (curve 1), θ_{20}(2, 4, 6), and $\theta_{1\Delta}$(3, 5, 7) on the magnetic field. The ratio J'/J is equal to 0.5 (2, 3), 0 (4, 5), and −0.5 (6, 7), g'/g = 0.606, S'/S = 1.

2. SPECTRUM OF IMPURITY OSCILLATIONS

The dynamic properties of a nonideal system are conveniently described with the aid of a Green's function defined by the equation[1)]

$$G_{nm}^{\alpha\beta}(\omega) = \lim_{s\to+0}\left\langle n,\alpha\left|\frac{1}{\omega - \hat{\mathcal{H}}_0 - \hat{V} - is}\right|m,\beta\right\rangle, \tag{10}$$

where α,β = 1, 2, 3, 4 and the wave functions $|n,\alpha\rangle$ determine the following states of the system:

$$\begin{aligned} |n,1\rangle &= a_{1n}^+|0\rangle, \quad |n,2\rangle = a_{1n}|0\rangle,\\ |n,3\rangle &= a_{2n}^+|0\rangle, \quad |n,4\rangle = a_{2n}|0\rangle; \end{aligned} \tag{11}$$

$|0\rangle$ is the wave function corresponding to the absence of elementary excitations in the antiferromagnet. From (10) we easily obtain the Dyson equation relating the Green's function $G(\omega)$ with the Green's function of an ideal crystal $G^0(\omega)$:

$$G(\omega) = G^0(\omega) + G^0(\omega)\hat{V}[1 - G^0(\omega)\hat{V}]^{-1}G^0(\omega), \tag{12}$$

from which we see that the poles of $G(\omega)$, which determine the energy spectrum of the impurity crystal, can be found from the equation

$$D(\omega) = \mathrm{Re\,Det}\,[1 - G^0(\omega)\hat{V}] = 0. \tag{13}$$

By virtue of the local character of the perturbation, the rank of the matrix (13) is finite (R = 36). However, since we take into account only the interaction between the different magnetic sublattices, the rank of the matrix is decreased by a factor of two.

The Green's function of an ideal crystal is obtained from (10) with $\hat{V}$ = 0:

$$\begin{aligned} G_{nm}^{011}(\omega) &= -G_{nm}^{022}(-\omega) = G_{nm}^{033}(\omega) = -G_{nm}^{044}(-\omega)\\ &= \frac{1}{N}\sum_k\{(\omega - 8JS)(\omega + 8JS)^2 + 64J^2S^2[(\omega + 8JS)\sin^4\theta\\ &\quad - (\omega - 8JS)\cos^4\theta]\varphi_k^2\}[(\omega^2 - \omega_{1k}^2)(\omega^2 - \omega_{2k}^2)]^{-1}e^{i(k,n-m)}, \end{aligned} \tag{14a}$$

$$\begin{aligned} G_{nm}^{012}(\omega) &= -G_{nm}^{021}(\omega) = G_{nm}^{034}(\omega) = -G_{nm}^{043}(\omega)\\ &= \frac{256J^3S^3}{N}\sin^2 2\theta\sum_k\frac{\varphi_k^2}{(\omega^2 - \omega_{1k}^2)(\omega^2 - \omega_{2k}^2)}e^{i(k,n-m)}, \end{aligned} \tag{14b}$$

$$\begin{aligned} G_{nm}^{013}(\omega) &= G_{nm}^{031}(\omega) = -G_{nm}^{024}(-\omega) = -G_{nm}^{042}(-\omega)\\ &= \frac{8JS\cos^2\theta}{N}\sum_k\frac{(\omega + 8JS)^2 - 64J^2S^2\cos 2\theta\varphi_k^2}{(\omega^2 - \omega_{1k}^2)(\omega^2 - \omega_{2k}^2)}\varphi_k e^{i(k,n-m)}, \end{aligned} \tag{14c}$$

$$\begin{aligned} G_{nm}^{014}(\omega) &= -G_{nm}^{041}(\omega) = -G_{nm}^{023}(\omega) = G_{nm}^{032}(\omega)\\ &= \frac{8JS\sin^2\theta}{N}\sum_k\frac{\omega^2 - 64J^2S^2(1 + \varphi_k^2\cos 2\theta)}{(\omega^2 - \omega_{1k}^2)(\omega^2 - \omega_{2k}^2)}\varphi_k e^{i(k,n-m)}. \end{aligned} \tag{14d}$$

Here

$$\varphi_k = \frac{1}{8}\sum_\Delta e^{ik\Delta} = \cos\frac{ak_x}{2}\cos\frac{ak_y}{2}\cos\frac{ak_z}{2}$$

(a is the period of the unit cell); ω_{1k} and ω_{2k} are the frequencies of the spin-wave excitations in an ideal crystal:

$$\begin{aligned} \omega_{1k}^2 &= 64J^2S^2(1 + 2\varphi_k\cos^2\theta + \varphi_k^2\cos 2\theta),\\ \omega_{2k}^2 &= 64J^2S^2(1 - 2\varphi_k\cos^2\theta + \varphi_k^2\cos 2\theta). \end{aligned} \tag{15}$$

It is seen from (15) that the second branch of the spin waves is nonactivational, and the first branch is activa-

[1)] We use throughout a system of units in which $\hbar$ = 1.

tional, and in addition, when $H > H_E/\sqrt{3}$ ($H_E = 16\ JS/\mu g$) the dispersion of the first branch changes from normal to anomalous.

The cubic symmetry of the crystal in question makes it possible to simplify (13) appreciably. To this end we introduce the unitary-transformation matrix

$$U = \frac{1}{\sqrt{8}} \begin{bmatrix} \sqrt{8} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 1 & 1 & 1 & 1 & 1 & 1 & 1 \\ 0 & 1 & -1 & 1 & -1 & 1 & -1 & 1 & -1 \\ 0 & 1 & -1 & -1 & 1 & -1 & -1 & 1 & 1 \\ 0 & 1 & 1 & -1 & -1 & -1 & 1 & 1 & -1 \\ 0 & 1 & -1 & -1 & 1 & 1 & 1 & -1 & -1 \\ 0 & 1 & 1 & -1 & -1 & 1 & -1 & -1 & 1 \\ 0 & 1 & 1 & 1 & 1 & -1 & -1 & -1 & -1 \\ 0 & 1 & -1 & 1 & -1 & -1 & 1 & -1 & 1 \end{bmatrix} \quad (16)$$

which enables us to change from the basis of the vectors $|\mathbf{n}, \alpha\rangle$ to a new wave-function basis that breaks up the 18-dimensional space into eight orthogonal subspaces, each of which is transformed in accordance with one of the irreducible representations of the point group O_h. We shall designate them arbitrarily as s, p, d, f. The determinant (13) breaks up in the new basis into the following product:

$$D(\omega) = D_s(\omega) D_p^3(\omega) D_d^3(\omega) D_f(\omega), \quad (17)$$

from which we see that the p and d levels are triply degenerate, with D_S, D_p, D_d, and D_f taking the form

$$D_s = \begin{bmatrix} 1 - 8(v_1 G_{00}^{011} + v_3 G_{01}^{031} - v_4 G_{01}^{032}) & 8(v_1 G_{00}^{012} + v_3 G_{01}^{032} - v_4 G_{01}^{031}) \\ -8(v_1 G_{00}^{021} + v_3 G_{01}^{041} - v_4 G_{01}^{042}) & 1 + 8(v_1 G_{00}^{022} + v_3 G_{01}^{042} - v_4 G_{01}^{041}) \\ -\sqrt{8}(8v_1 G_{01}^{013} + v_3 A_s^{11} - v_4 A_s^{12}) & \sqrt{8}(8v_1 G_{01}^{024} + v_3 A_s^{12} - v_4 A_s^{11}) \\ -\sqrt{8}(8v_1 G_{01}^{023} + v_3 A_s^{21} - v_4 A_s^{22}) & \sqrt{8}(8v_1 G_{01}^{024} + v_3 A_s^{22} - v_4 A_s^{21}) \end{bmatrix}$$

$$\begin{bmatrix} -\sqrt{8}(v_2 G_{01}^{031} + v_3 G_{00}^{011} - v_4 G_{00}^{012}) & \sqrt{8}(v_2 G_{01}^{032} + v_3 G_{00}^{012} - v_4 G_{00}^{011}) \\ -\sqrt{8}(v_2 G_{01}^{041} + v_3 G_{00}^{021} - v_4 G_{00}^{022}) & \sqrt{8}(v_2 G_{01}^{042} + v_3 G_{00}^{022} - v_4 G_{09}^{021}) \\ 1 - (v_2 A_s^{11} + 8v_3 G_{01}^{013} - 8v_4 G_{01}^{014}) & (v_2 A_s^{12} + 8v_3 G_{01}^{014} - 8v_4 G_{01}^{013}) \\ -(v_2 A_s^{21} + 8v_3 G_{01}^{023} - 8v_4 G_{01}^{024}) & 1 + (v_2 A_s^{22} + 8v_3 G_{01}^{024} - 8v_4 G_{01}^{023}) \end{bmatrix}, \quad (18)$$

$$D_i(\omega) = \begin{bmatrix} 1 - v_2 A_i^{11} & v_2 A_i^{12} \\ -v_2 A_i^{21} & 1 + v_2 A_i^{22} \end{bmatrix}, \quad i = p, d, f, \quad (19)$$

where

$$A_s = G_{00}^0 + 3G_{12}^0 + 3G_{13}^0 + G_{17}^0, \quad A_p = G_{00}^0 - G_{12}^0 - G_{13}^0 + G_{17}^0,$$
$$A_d = G_{00}^0 + G_{12}^0 - G_{13}^0 - G_{17}^0, \quad A_f = G_{00}^0 - 3G_{12}^0 + 3G_{13}^0 - G_{17}^0. \quad (20)$$

Using a computer, we calculated the Green's functions of an ideal crystal G_{00}^0, G_{12}^0, G_{13}^0, and G_{17}^0, and we also determined the solutions of the equations $D_i(\omega) = 0$ for $\cos\theta$ varying from 0 to 1 in steps of 0.1. The solution of the equation $D_S(\omega) = 0$ as a function of the magnetic field H is shown in Fig. 3 for the following values of the impurity parameters: $J'/J = -0.5$; -0.1; 0; 0.1; $g'/g = 0.606$; $S'/S = 1$. Figure 4 shows the dependence of the d and f levels on the magnetic field at $J'/J = 0$; $g'/g = 0.606$; $S'/S = 1$.

The greatest interest attaches to local oscillations of the s type since, as shown by the analysis, they are the only ones contributing to the high-frequency magnetic susceptibility $\chi(\omega)$. Figure 3 shows, in addition to the s levels, also a plot (dashed) of the frequency of the homogeneous AFMR as a function of the magnetic field,

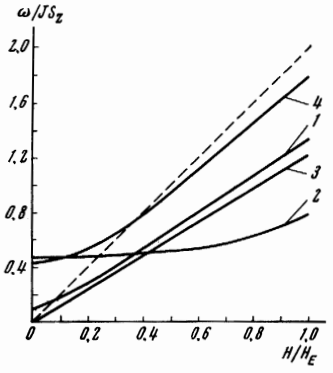

FIG. 3. Dependence of s levels on the magnetic field: the ratio J'/J is equal to −0.1 (curve 1), 0.5 (2), 0 (3), −0.5 (4), g'/g = 0.606, S'/S = 1.

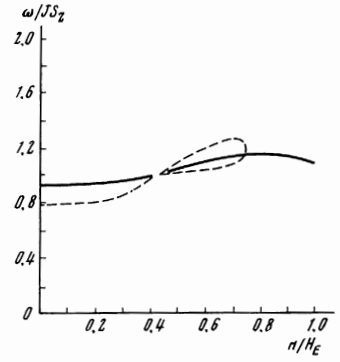

FIG. 4. Dependence of f levels (solid line) and d levels (dashed) on the magnetic field: J'/J = 0, g'/g = 0.606, S'/S = 1.

from which we see that in certain magnetic fields (points of intersection of the AFMR line with the s levels) there can occur resonant excitation of the local impurity oscillations by a homogeneous high-frequency field. This should become manifest experimentally in a splitting of the AFMR line, the value of the splitting being proportional to $\sqrt{c}$, where c is the concentration of the magnetic impurities. On the other hand, the levels of the p, d, and f types can be observed by investigating the inelastic single-quantum cross section for the scattering of slow neutrons by impurity antiferromagnets.

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