Exchange amplification and magnetic impurity oscillation (MIO) quenching in antiferromagnets (AF)

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The anomalously high intensity of the absorption line of Mn^{2+} in the antiferromagnet CoF_2 ($I_{\text{impurity}}/I_{\text{AFMR}} \sim 1$) at a very low Mn^{2+} concentration ($c \approx 0.01$–0.03%) has been observed experimentally and is explained theoretically. The amplification effect vanishes when the MIO fall between two (AFMR) frequencies.

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Cobalt fluoride (CoF_2, $T = 37\,^\circ K$, crystallographic symmetry D_{4h}^{14}) is the only known antiferromagnet in which the MIO frequency $\nu_{\text{imp}}(H=0)$ lies below the spin-wave band.[1,2] At $H > H^*$, the MIO fall in the spin-wave band. We investigated antiferromagnetic resonance (AFMR) in CoF_2 with small amounts of Mn^{2+} impurity ($c \approx 0.01$–0.03 wt.%)[1)]. The measurements were performed at wavelengths 270–400 μ, $0 < H < 100$ kOe($\mathbf{H} \parallel C_4$), and $T = 4.2\,^\circ K$.[3] We investigated the behavior of the position and intensity of the AFMR absorption lines (I_A) and of the Mn^{2+} impurity ($I_{\text{imp 1}}$). Figure 1 shows plots of the absorption lines for four wavelengths. Two minima are observed on curve 1. The minimum in the field near 20 kOe corresponds to the MIO, while the minimum near 55 Oe corresponds to AFMR. As seen from this curve, the MIO intensity is only 30–40% smaller than the AFMR intensity, in spite of the fact that the Mn^{2+} concentration is extremely small. This is surprising, inasmuch as $I_{\text{imp}} \sim c$. Figure 2 shows plots of the AFMR frequencies ν_{A1} and ν_{A2} and of the impurity frequencies ($\nu_{\text{imp 1}}$ and $\nu_{\text{imp 2}}$) against the magnetic field. In a magnetic field $H^* = 49$ kOe and at a frequency $\nu = 918 \times 10^9$ sec^{-1} ($\lambda = 326.8$), the branches intersect, and at $H > H^*$ the frequency $\nu_{\text{imp 1}}$ falls in the spin-wave band. Curve 2 of Fig. 1 corresponds to a frequency $\nu < \nu^*$, but closer to the point of intersection in comparison with curve 1. We see that $I_{\text{imp 1}}$ increases as $\nu \to \nu^*$ and as $H \to H^*$. The frequency $\nu_{\text{imp 1}}(H)$ is not observed at all at $\nu > \nu^*$, although it does manifest itself in a certain asymmetry of curve 3 of Fig. 1. In still stronger magnetic fields (curve 4), the AFMR line becomes symmetrical. To understand the principal features of the phenomenon, let us consider small MIO in an antiferromagnetic matrix, disregarding the interaction of the impurities with one another. Let the impurity magnetic moment $\vec{\mu}_0$ be located at a site numbered "0" and let it be directed along $\mathbf{H} = \{0, 0, H\}$. This corresponds to a linear $\nu_{\text{imp 1}}(H)$ dependence. Then the exchange interaction between the impurity and the matrix can be written in the form

$$\vec{\mu}_0 \sum_i \sum_{\vec{\delta}_i} J_{0i}\, \mathbf{S}_{\vec{\delta}_i}\,, \qquad (1)$$

where J_{0i} are the exchange integrals of the impurity with the matrix ions located in the i-th coordination sphere (CS), $\mathbf{S}_{\vec{\delta}_i}$ are the magnetic moments of the matrix ions located in one and the same CS, Σ_i is the sum over all the CS, and $\Sigma_{\vec{\delta}_i}$ is the sum over the magnetic moments of the corresponding CS. Considering only homogeneous matrix oscillations corresponding to AFMR, when all the magnetic moments in the same sublattice remain parallel in the course of the motion, and disregarding the "spin cancellations" in the system, we can, on the basis of (1), write down the thermodynamic potential for one impurity magnetic moment in the exchange approximation in a magnetic field

$$f_{\Pi} = \frac{1}{N}\,[\,J_2\, \vec{\mu}_0 \mathbf{M}_2 + J_1\, \vec{\mu}_0 \mathbf{M}_1\,] - \vec{\mu}_0 \mathbf{H}, \qquad (2)$$

where M_1 and M_2 are the magnetizations per unit volume of the corresponding sublattices of the matrix, and N is the number of magnetic ions in one sublattice per unit volume. Writing down the thermodynamic potential of the matrix in standard form

$$\Phi = \frac{B}{2}\mathbf{M}^2 + \frac{a}{2}L_z^2 + \frac{b}{2}M_z^2 + d\,(M_x L_y + M_y L_x) - \mathbf{M}\mathbf{H} \qquad (3)$$

and considering small oscillations of the impurity in the matrix,[4,5] we can show that the impurity oscillations are excited by an effective field $h_{\text{eff}} = h + h_{\text{exch}}$. The gain η depends both on the matrix parameters and on the coupling between the matrix and the impurity

$$\eta = 1 + (\gamma_A/\gamma_{\Pi})\, \nu_{\Pi}(H=0)\, \gamma_A \sqrt{H_A(H_A + 2H_E)}\,[\,(1 - K^+)/(\nu^2 - \nu_{A1}^2)$$
$$- (1 - K^-)/(\nu^2 - \nu_{A2}^2)\,], \qquad (4)$$

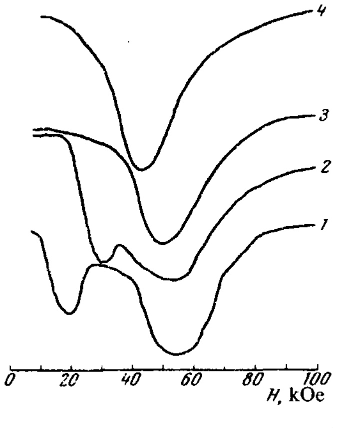

FIG. 1. Resonance-absorption lines at the wavelengths $\lambda = 340$, 336.2, 325.4, and 317.8 (1 to 4, respectively).

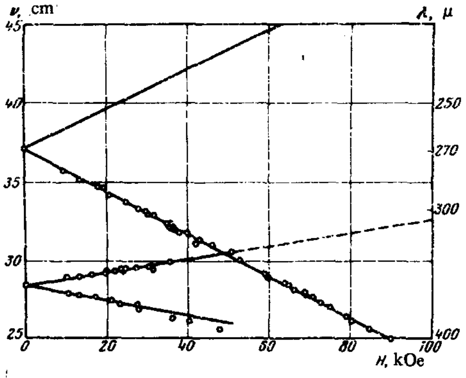

FIG. 2a. Dependence of the AFMR frequencies (1—ν_{A1} and 2—ν_{A2}) and of the impurity frequencies (3—$\nu_{\text{imp }1}$, 4—$\nu_{\text{imp }2}$) on the magnetic field.

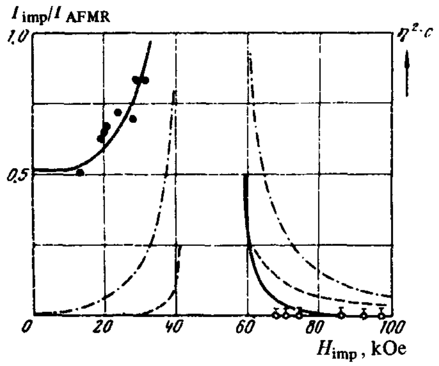

FIG. 2b. Dependence of $I_{\text{imp }1}/I_{A2}$ on the magnetic field. The solid line is drawn in accordance with formulas (4) and (5); the dashed and dash-dot lines are calculated with $J_2 = 0$ and $J_1 = 0$, respectively.

$$K^{\pm} = \sqrt{H_A/(H_A + 2H_E)}$$
$$\times\left\{\left[(J_1 + J_2)/(J_2 - J_1)\right]\left[1 \pm H/\sqrt{H_A(H_A + 2H_E)}\right] - H/H_A\right\} \qquad (5)$$

where γ_A and γ_{imp} are the gamma-factors of the matrix and impurity, $\nu_{A1,2} = (\sqrt{H_A(H_A+2H_E)} \pm H)^2 - H_D^2$, $H_A = -2aM$, $H_E = (B-A)M$, and $H_D = 2dM$. The calculation was carried out without taking into account the interaction of the oscillations near H^* and is valid for antiferromagnetism of the easy-axis type, including the Dzyaloshinskiĭ interaction. The solid line in Fig. 2b is the calculated plot of $c\eta^2 \sim I_{\text{imp }1}$ for CoF$_2$, where $H_A = 120$ kOe, $H_E = 400$ kOe, $H_D = 180$ kOe, [1,6] and $J_2/J_1 = 1.4$. The points on Fig. 2b are experimental. To understand the physical picture of the phenomenon, we shall assume that $K^+ \approx K^- = K = \sqrt{H_A/(H_A+2H_E)}(J_2+J_1)/(J_2-J_1)$. Then the observed amplification of the oscillations at $H<H^*$ and the damping at $H>H^*$ will occur when $|K|>1$. This can happen only if $H_A/(H_A+2H_E)$ is not small and there is no large difference between J_1 and J_2 (J_1 and J_2 are of the same sign). The parameter $H_A/(H_A+2H_E)$ determines the dynamic angle between $\mathbf{M}_1$ and $\mathbf{M}_2$ in a rotating coordinate system, and smallness of $J_2 - J_1$ corresponds to a large high-frequency susceptibility of the impurity and a small ν_{imp}. The observed effect differs in principle from the known enhancement of the field at the nuclei in magnets, in that the presence of two matrix sublattices is required in principle in our case (see Fig. 2b). At $H>H^*$ the frequency $\nu_{\text{imp }1}$ falls between ν_{A1} and ν_{A2}, and the high-frequency fields are subtracted from the two normal oscillations ν_{A1} and ν_{A2}, so that damping is observed. It is possible that this mechanism caused the failure to observe MIO in electromagnetically-excitated antiferromagnets, particularly in the investigation of AFMR in CoCO$_3$.[7]

It follows from our experiment that $J_2/J_1 = 1.1$ to 1.5 for CoF$_2$.

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