De Haas-van Alphen effect in the band antiferromagnet $FeGe_2$: Development of spin splitting

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The de Haas-van Alphen (dHvA) effect in the antiferromagnetic compound $FeGe_2$ is studied experimentally. A sharp suppression of the amplitude of the first harmonic of the dHvA oscillations is observed for a number of orbits on the Fermi surface of $FeGe_2$. The change in the conditions for suppression of the amplitude ("spin zeroes") when $FeGe_2$ is doped with cobalt is determined. A method for analyzing the experimental data is proposed in which the g-factors of the conduction electrons are computed on the basis of calculations of the spin-polarized electron structure of the band antiferromagnet taking spin-orbital interactions into account. The exchange enhancement factor for spin paramagnetism in $FeGe_2$ is also calculated. © *2014 AIP Publishing LLC*.
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The discovery of superconductivity in a number of layered compounds of iron with sp-elements (As, Se, Te), in which anitferromagnetic (AFM) ordering is also realized, is of interest for research on the nature and parameters of electronic interactions in these (not necessarily superconducting) systems at a microscopic level. Iron digermanide $FeGe_2$ is a representative of this class of compounds. $FeGe_2$ has a tetragonal structure with alternating layers of iron and germanium and is an antiferromagnet in its ground state, where magnetic ordering is provided by conduction electrons. The chemical and magnetic lattices of $FeGe_2$ coincide,[1] and this simplifies theoretical analysis of its electronic properties.

The de Haas-van Alphen (dHvA) effect is a powerful tool for studying the electronic states on the Fermi surface. It can be used to determine the areas of the extremal cross sections of the Fermi surface, as well as the characteristics of the interactions of charge carriers on extremal trajectories.[2–4] The dHvA effect in $FeGe_2$ was first observed by Verkin *et al*.[5] The electronic spectrum of the paramagnetic and magnetically ordered phases of $FeGe_2$ have been calculated.[6–8] These calculations show that the minimum total energy $FeGe_2$ corresponds to a collinear AFM structure with alternating (100) ferromagnetic layers of the moments of iron with $m = 1.24\mu_B$, in accordance with neutron diffraction measurements.[1] These studies have confirmed the existence of a sheet of a hole Fermi surface similar to an octahedron slightly stretched out along a tetragonal axis. This sheet of the Fermi surface centered at the Γ point of the Brillouin band is also responsible for quantization of the magnetization oscillations in $FeGe_2$ that are the subject of this paper.

We have studied the dHvA effect in single crystals of $FeGe_2$, as well as in a sample of cobalt-doped $FeGe_2$ (1000 ppm Co) at a minimum temperature of 1.7 K and in magnetic fields up to 8 T. When the magnetic field is directed near the [001] axis (i.e., for extremal trajectories near the basis plane of the octahedron) a sharp suppression of the amplitude of the first harmonic of the oscillations has been observed; this is caused by spin splitting of the Landau levels and a special relationship between the magnitudes of the spin and orbital splittings. Experimental and theoretical studies of spin splitting in the electronic spectrum offer a fundamental possibility of gaining information on the interactions among the conduction electrons.[4]

According to the Lifshits-Kosevich theory,[2,3] because of the superposition of the oscillations from electrons with two spin directions the resulting amplitude of the first harmonic of the dHvA oscillations is proportional to the spin factor

$$M \sim \cos[(\pi/2)g_c m_c/m_0], \tag{1}$$

where m_c is the cyclotron mass for a given orbit on the Fermi surface, m_0 is the free electron mass, and g_c is the orbital g-factor. The condition for suppression of the amplitude (the so-called spin-splitting zero (SSZ)) follows from Eq. (1):

$$g_c m_c/m_0 = 1, 3, \ldots. \tag{2}$$

Detection of the SSZ for a given direction of the magnetic field makes it possible immediately to find the effective g-factor for the corresponding orbit from the experimentally determined electron (hole) cyclotron mass, without having to rely on a more cumbersome, less accurate, and not always unique analysis of the amplitude. It should be noted that the g-factors determined from dHvA experiments have been renormalized by the multiparticle exchange interaction:[4] $g_c = Sg_b$, where g_b are the "band" g-factors which can be determined from calculations of the band structure and S is the Stoner enhancement factor.

The effective cyclotron masses of the "octahedron" in $FeGe_2$ for fields directed along the principal crystallographic axes are[7] m_c/m_0 [001] = 0.51 and m_c/m_0 [100] = m_c/m_0

[110] = 0.40, and for values of the g-factor close to $g_0 = 2$ for free electrons, these are indeed favorable to the conditions for SSZ according to Eq. (2) ($g_c m_c/m_0 = 1$) when the field deviates from the [001] axis by some angle θ. Since the orbital g-factors can differ significantly from g_0 owing to exchange enhancement and can have substantial anisotropy because of spin-orbital interactions,[4] in this paper we search for manifestations of SSZ in $FeGe_2$ for different directions of the magnetic field.

Deep amplitude drops of the type in SSZ have also been detected in the neighborhood of the [100] axis (Fig. 1). The main feature of the observed zero spins in $FeGe_2$ (pure and doped with cobalt) is that their position depends on the magnitude of the magnetic field (see Fig. 1). In addition, the amplitude of the oscillations right at the SSZ points does not go strictly to zero, but remains finite, even if low. In particular, this amplitude is sufficient for determining the effective masses directly at the SSZ (we retain this term for the "incomplete" spin zeroes) around the [100] axis. To within the limits of accuracy, they coincide with the measured value of $m/m_0[100] = 0.40$. Figure 2 shows some stereographic projections of the profiles of SSZ_{100} around the [100] axis with different values of the magnetic field for pure $FeGe_2$ and $FeGe_2$ + 1000 ppm Co. Only reliably resolved amplitude minima are shown, for which a detailed analysis of scattering effects is needed to reveal the SSZ.

The SSZ_{100} profile is stretched out in the [001] direction and broadens as the field is increased, at least in this direction in pure $FeGe_2$ and almost isotropically in $Fe(Co)Ge_2$. The shift in the profiles with increasing field is small, about 2° in the working range (Fig. 2), so it is not possible to establish a functional relationship between the field and the angular shift of SSZ_{100}. The Co impurity had little effect on the position of the spin zero profile relative to the [100] axis and its field dependence (at least in the (100) plane).

The influence of the field and Co impurity on the SSZ_{001} profile around the tetragonal axis is considerably stronger; here these factors bring the profile closer to the [001] axis. The relation of the field $H_{SSZ}(\theta)$ to the angle θ between direction of the field and the [001] axis in the (110) plane can be well approximated by the empirical formula

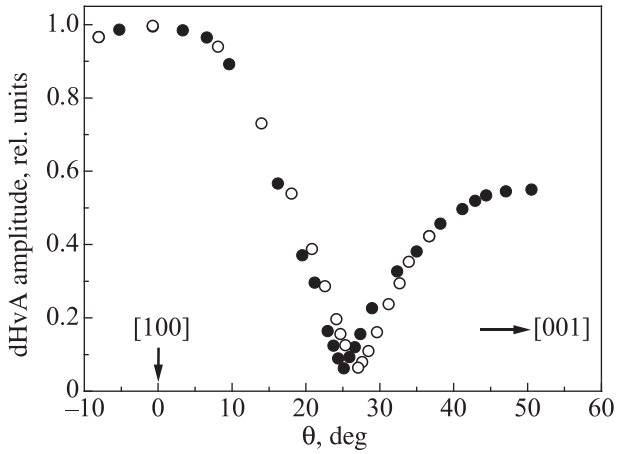

FIG. 1. Angular dependence of the amplitude of the oscillations in the magnetic moment of $FeGe_2$ in the vicinity of SSZ_{100} "spin zeroes" for different magnetic fields H (T): 4 (●) and 6.5 (○).

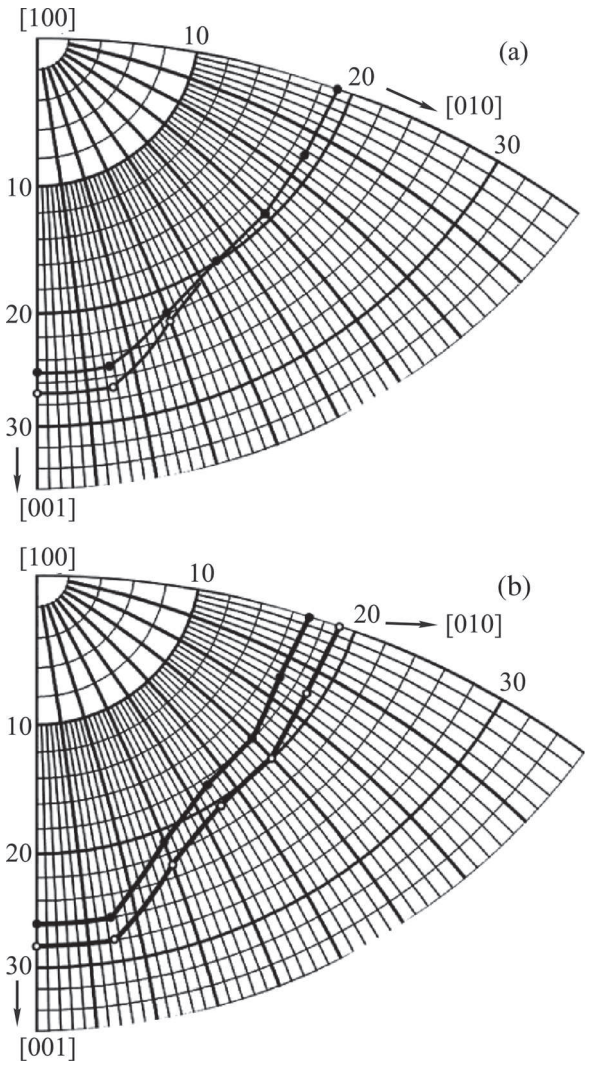

FIG. 2. Stereographic projections of SSZ_{100} profiles for samples of $FeGe_2$ (a) and $FeGe_2$ + 1000 ppm Co (b) in different fields H (T): (a) 4 (●), 6.5 (○); (b) 3.5 (●), 6.5 (○).

$$H_{SSZ}(0) - H_{SSZ}(\theta) = k(1 - \cos\theta), \qquad (3)$$

with the right part essentially being the same for $FeGe_2$ and the alloy. The field $H_{SSZ}(0)$ at which the profile SSZ_{001} should be stretched toward a point on the [001] axis (9.8 T in $FeGe_2$ and 7.9 T in the alloy) and the coefficient $k = 138$ T were obtained by fitting Eq. (3) to the experimental data (Fig. 3). Adding 1000 ppm of the Co impurity lowers the field H_{SSZ} by roughly 2 T, without changing its angular dependence. Thus, this dependence is intrinsic to the FeGe2 matrix and is not caused by deviations from stoichiometry or some other structural defects. According to a linear extrapolation, 4000 ppm Co is sufficient to shift SSZ_{001} out of the observation zone.

SSZ depends on the field mainly because of a desynchronization of the spin and orbital splitting of the levels in the field. This may be caused by a field-independent interaction which adds to the spin splitting because of an exchange interaction of the band electrons with localized moments of impurity atoms (in particular, in Kondo systems[4]) or with ordered magnetic moments in band antiferromagnets.[9] Then the way the scattering of the electrons depends on spin orientation leads to a difference in their contributions to the

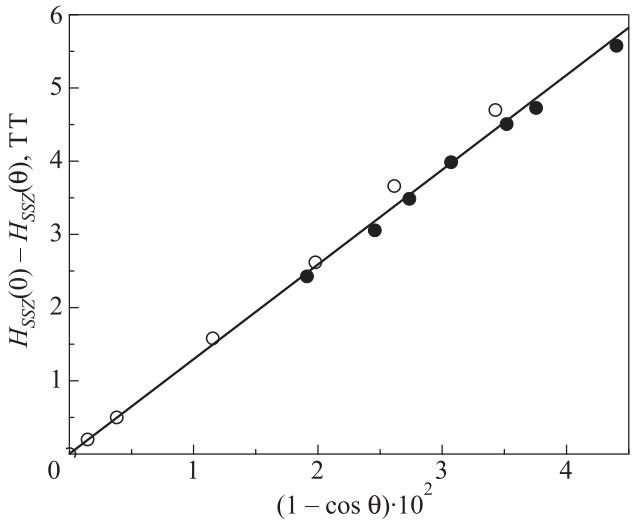

FIG. 3. Approximating the experimental angular dependences $H_{SSZ}(0)$-$H_{SSZ}(\theta)$ for $FeGe_2$ (●) and $FeGe_2$ + 1000 ppm Co (○) by Eq. (3) in the vicinity of the [001] axis.

amplitude of the oscillations and to its incomplete suppression at the "spin zeroes."

In the case of dominant exchange with impurity moments, the condition for realization of SSZ depends on the field and Eq. (2) is modified in the following way:[4]

$$(g_c - H_{\rm ex}/H)m_c/m_0 = 1, 3, \ldots. \qquad (4)$$

Here antiferromagnetic exchange is assumed and $H_{\rm ex}$ is the level exchange splitting parameter, which is proportional to the magnetic impurity concentration and to the exchange integral.

The conditions for SSZ in band antiferromagnets with coincident periods of their magnetic and chemical lattices have been examined[9] using perturbation theory to account for the influence of the Coulomb and exchange potentials. The results, expressed in terms of the band characteristics, predict a field dependence for SSZ. It turns out that numerical calculations for $FeGe_2$ based on that theory[9] are inappropriate because the simplified model of the spectrum upon which the theory is based does not agree with that of real compounds of transition *d*-metals.

The simplicity of an octahedral Fermi surface of $FeGe_2$ is, in fact, illusory. This is evident from the disagreement in the anisotropy of the cross sections of the surface and in the effective masses: large masses are observed on the thin "waist." Calculations of the spectrum $E(k)$ show that the Fermi surface of $FeGe_2$ is characterized by strong hybridization of the *d*-states of iron and the *p*-states of germanium. This is confirmed by calculations[7] that reveal a great sensitivity of the shape and dimensions of the octahedral surface and of the effective masses to the type of AFM ordering. Not every sheet of the Fermi surface is suitable for testing of the magnetic structure. Recent studies of the dHvA effect in the antiferromagnetic compound CrB_2 show[10] that the Fermi surface observed in it is created by p-states of boron with no dependence whatever on the magnetic ordering.

The dHvA "spin zeroes" in $FeGe_2$ observed here can be used to determine the corresponding orbital *g*-factors of the conduction electrons, which depend on features of the spin-orbital, electron-electron, and exchange interactions in the antiferromagnetic $FeGe_2$. Measurements and theoretical analysis of the g-factors make it possible, in principle, to extract information on the features of the electronic structure and multiparticle interactions on the Fermi surface. However, the methods for calculating the *g*-factors in metals are still one of the least developed methods in band theory, especially for band (itinerant) AFM systems.

An original method has been proposed[11] for self-consistent calculation of the spin-polarized electronic structure of metallic systems in an external magnetic field in terms of the density functional theory (DFT) and the linear muffin-tin orbital (LMTO) formalism with the full potential[12] by including the Zeeman operator $\mu_B H(2\hat{S} + \hat{L})$, where $\hat{S}$ is the spin operator and $\hat{L}$ is the orbital angular momentum operator, in the hamiltonian. The present calculations of the induced spin and orbital magnetic moments in an external field of 10 T yielded the corresponding components of the magnetic susceptibility tensor χ in the AFM phase of $FeGe_2$. The van Vleck orbital contribution to χ was significant (about 40% of the total paramagnetic susceptibility), while the exchange enhancement of the spin paramagnetism (which shows up in the *g*-factors through the Stoner factor *S*) was relatively small at $S = 1.7$. This is explained by the location of the Fermi level in the vicinity of a deep minimum in the density of states, in accord with the partial "dielectrization" of the spectrum owing to the appearance of AFM "gaps" in a number of directions of the Brillouin band.[7]

In accordance with the band theory of spin polarized electronic spectra, in a magnetic field *H* the spin degeneracy of the electronic states in the conduction band is removed and each level splits into two with energies equal to

$$E_n(\mathbf{k})^+ - E_n(\mathbf{k})^- = g_n^\alpha(\mathbf{k})\mu_B H, \qquad (5)$$

where *n* denotes the number of the energy band and α, the direction of the magnetic field. Thus, according to Eq. (5), by direct computation of the splittings of the energy levels in a magnetic field in the formalism of Refs. 11 and 12, it is possible to calculate the anisotropic *g*-factors on the Fermi surface of AFM $FeGe_2$. Here the effects of spin-orbital, electron-electron, and AFM exchange interactions will be taken into account through a unified self-consistent DFT first-principles calculation of the spectrum. Detailed calculations of the orbital g-factors on the Fermi surface of AFM $FeGe_2$ and a corresponding analysis of the behavior of the "spin zeroes" obtained in dHvA experiments will be the subject of a separate study and go beyond the scope of the present paper. The experimental data shown here offer the possibility for study of a rather wide set of extremal orbits with a fixed ratio of the spin and orbital splittings of the energy levels, including ones corresponding to charge carrier motion in crystalline layers of the same chemical nature.

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