PHYSICAL REVIEW B, VOLUME 65, 064205

Electron spin resonance in an icosahedral $Al_{63}Cu_{25}Fe_{12}$ quasicrystal

J. Dolinšek, D. Arčon, A. Zorko, and M. Klanjšek
J. Stefan Institute, University of Ljubljana, Jamova 39, SI-1000 Ljubljana, Slovenia

C. Saylor and L. C. Brunel
National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida

P. Brunet and J. M. Dubois
Laboratoire de Science et Génie des Matériaux et de Métallurgie (Unité Mixte de Recherche 7584), Centre d'Ingénierie des Matériaux, Ecole des Mines, Parc de Saurupt, F-54042 Nancy Cedex, France

(Received 29 May 2001; published 22 January 2002)

An electron spin resonance (ESR) study of an icosahedral $Al_{63}Cu_{25}Fe_{12}$ quasicrystal is presented. The ESR signal originates from two kinds of electronic spins—the itinerant conduction electrons and the localized *d* moments of magnetic Fe atoms. The ESR spectra are very broad, extending over up to 5 T in the magnetic-field-sweep experiment. The ESR signal could be detected by a high-field transmission ESR spectrometer operating at a frequency 109.270 GHz, whereas the too-small field sweep of 0–1 T prevented the ESR signal of the $Al_{63}Cu_{25}Fe_{12}$ quasicrystal to be fully observed by the conventional *X*-band reflection spectrometer. The ESR spectra are compared to the NMR spectra of the same sample. Both spectra are extremely broad and the origin of this large broadening is found to be similar in both cases. It results from the distribution of local electric fields that are present in quasicrystals due to the lack of translation periodicity of the quasiperiodic lattice and from the *s*-*d* exchange interaction between the conduction electrons and the localized Fe *d* moments. An anomalous temperature dependence of the ESR linewidth that broadens on cooling above 20 K, but narrows from 20 to 5 K by a factor 3.7, was detected. General aspects of the ESR spectroscopy in quasicrystals are also discussed.

DOI: 10.1103/PhysRevB.65.064205 PACS number(s): 61.44.Br, 71.23.Ft, 76.30.−v

I. INTRODUCTION

Magnetic resonance techniques provide an important tool for experimental investigations of physical properties of quasicrystals (QC's)—nonperiodic long-range ordered solids that obey forbidden symmetries of 5-, 8-, 10-, and 12-fold rotation axes. Experimental investigations of QC's with nuclear magnetic resonance (NMR) spectroscopy are numerous. NMR spectra of QC's are very broad, extending over several megahertz frequency scale, so that special techniques of field and frequency sweep have to be employed in order to record a quasicrystalline spectrum. Measurements of the static NMR line shape enable a study of the distribution of physically inequivalent lattice sites via a determination of the distribution of local electric field gradient (EFG) tensors.[1,2] The intrinsic magnetism of QC's containing transition-metal elements, such as Mn and Fe, can be studied via the temperature-dependent NMR linewidth and its frequency shift.[3,4] NMR diffusion measurements[5] enable detection of slow atomic motions with kilohertz frequencies, whereas the two-dimensional exchange NMR technique extends the observation window to ultraslow motions in the subhertz regime.[6] NMR spin-lattice relaxation also provides detailed information on the shape and magnitude of the electronic density of states[7] (DOS) of QC's in the vicinity of the Fermi level E_F.

Experimental studies of QC's with another kind of magnetic resonance technique—the nuclear quadrupole resonance (NQR)—are at present still scarce. Zero-field NQR was applied so far only to Al-Cu-Fe and Al-Cu-Ru powders[1] by measuring ^{27}Al NQR signals. NQR is insensitive to the orientation of the local EFG tensors, but detects the distribution of the EFG tensor eigenvalues that results from the distribution of local atomic environments around the resonating nuclei in the nonperiodic QC lattice.

The ^{27}Al NQR resonance lines were found strongly inhomogeneously broadened (the width of several megahertz) and centered at the frequency of about 4 MHz. The large width of the spectrum and the short spin-spin relaxation time T_2 encountered in QC's introduce a sensitivity problem, so that the NQR lines could be detected at low temperatures only, using a small additional Zeeman field to decouple the ^{27}Al nuclear spins, making T_2 longer. Due to these problems, the NQR method is less convenient for the investigations of QC's.

QC's exhibit very interesting electronic properties. Despite being alloys of metallic elements, their electrical resistivity generally exhibits a negative temperature coefficient, just opposite to that of regular metals. Their electronic DOS at the Fermi level exhibits a pseudogap, which in some cases shows the proximity of a metal-to-insulator transition at low temperatures.[8] It is believed that the increasing resistivity upon cooling and the metal-to-insulator transition occur due to the specific behavior of conduction electrons that undergo a gradual localization at low temperatures. The electrons occupy ‘‘critical’’ states that are neither extended in space nor localized on the atomic scale, but exhibit localization on an intermediate scale over many interatomic distances. The physics of conduction electrons thus represents a fundamental issue of study in QC's. There exists another interesting phenomenon emerging from localized unpaired electronic spins that are present in magnetic QC's, either containing transition metals (Mn, Fe) or rare-earth-metal atoms. The magnetic exchange interaction between the unpaired *d* of *f* moments leads to magnetic ordering phenomena of the spin glass or antiferromagnetic type at low temperatures. In view of that it would be highly desirable to perform studies of QC's by the method of electron spin resonance (ESR) that in principle detects signals from both the itinerant conduction electrons and the localized electronic spins. However, so far only QC's exhibiting ferromagnetism [Al-Mn-Si (Ref. 9) and Al-Mn-Pd-B (Ref. 10)] were investigated by ESR, where the existence of ferromagnetic resonances was reported in the ferromagnetic phase. It is astonishing that up until now—to the best of our knowledge—not a single ESR study was reported in literature on QC's that do not exhibit ferromagnetism. In this paper we report the ESR study of a prototypical high-quality icosahedral quasicrystal, namely the $Al_{63}Cu_{25}Fe_{12}$ compound. We discuss the general aspects of the ESR experiment on QC's and outline the experimental difficulties encountered due to the significant metallic character of the quasicrystalline samples in connection with the nonperiodic long-range order of the quasicrystalline lattice.

II. ESR IN NONMAGNETIC QUASICRYSTALS

The electrical resistivities of QC's are several orders of magnitude larger from those of regular metals. Typical resistivities below room temperature are in the range $\rho \approx 10^2 - 10^3\ \mu\Omega\,\mathrm{cm}$ for decagonal QC's and $\rho \approx 10^3 - 10^6\ \mu\Omega\,\mathrm{cm}$ for icosahedral QC's. This is to be contrasted with the resistivity of metallic aluminum where $\rho_{77\,\mathrm{K}} = 0.3\ \mu\Omega\,\mathrm{cm}$. However, the resistivities of QC's remain many orders of magnitude smaller from those of true insulators, so that QC's can generally be considered as weak metals (or semimetals). This metallic character represents one of the problems in ESR spectroscopy on QC's, as the electron spin resonance of conduction electrons is generally difficult to observe. Unlike NMR in metals, where the NMR signal can be practically always detected, ESR of conduction electrons was observed so far only on a limited number of metals. The reason is that the conduction-electron resonance linewidth is in many cases so large that the signal is unobservable. The weak ESR sensitivity of metals originates from two reasons. First is the fact that the intensity of the ESR line is proportional to the static spin susceptibility that, for conduction electrons, is of a Pauli type, its order of magnitude being given by $\chi_P \approx (g\mu_B)^2/k_B T_F$. Here μ_B is the Bohr magneton, g the electronic g factor and $T_F = E_F/k_B$ is the Fermi temperature (of the order 10^4 K). This susceptibility remains four orders of magnitude smaller than the Curie-type paramagnetic susceptibility of localized electrons at temperatures close to liquid He for a comparable density of electronic spins (provided the localized electrons are not in a magnetically ordered state). The second reason for the weakness of the ESR signal is the skin effect, where the microwave field penetrates into the metallic sample only up to the distance of the order of a skin depth $\delta = \sqrt{\rho/\mu_0 \pi \nu}$ (with μ_0 being the permeability of a vacuum). The skin effect drastically reduces the number of excited electrons that contribute to the ESR signal. In *X*-band ESR with the resonance frequency of free $(g_F \approx 2)$ electrons at $\nu = 9.6$ GHz, a typical resistivity of a QC sample $\rho = 2000\ \mu\Omega\,\mathrm{cm}$ results in a skin depth of $\delta = 23\ \mu\mathrm{m}$. In high-field ESR performed at $\nu = 109$ GHz, the skin depth reduces to $\delta = 7\ \mu\mathrm{m}$. The microwave field can thus excite a tiny amount of conduction electrons within the surface layer of the sample only.

The g factor of conduction electrons in metals[11] is generally not very different from the free-electron value $g_F \approx 2$. The reason is that the exchange interaction between the fast-moving conduction electrons is averaged to zero and thus unable to change the resonance frequency. The small difference between the actual- and the free-electron g values of the order $g - g_F \leq 10^{-2}$ found in regular metals is attributed to the spin-orbit coupling. A related situation should apply to QC's too. There the average g value of conduction electrons should again be centered close to the free-electron value for the same reason. The spin-orbit coupling, however, deserves more attention. NMR measurements[2] of the static line shape have shown the existence of a very strong inhomogeneous broadening of electric quadrupolar origin. The broadening is induced by a broad distribution of crystalline electric fields (CEF's) in the quasiperiodic lattice, which result in quenching of the electronic orbital motion ($\vec{L} = \mathbf{0}$) in the absence of an external magnetic field. In the field the orbital motion becomes partially unquenched, leading to g shifts via the spin-orbit coupling. The spin-orbit coupling constant is proportional to the electric field in which the electron is moving. A large distribution of CEF's is expected to introduce a large distribution of g shifts that should result in broad ESR spectra, in analogy to the extremely broad NMR spectra of QC's. We can anticipate that the underlying reason for the very broad spectra in NMR and ESR experiments on QC's should be similar—the nonperiodic quasicrystalline lattice generates a large number of different local atomic environments that introduce a broad distribution of local electric fields.

In regular metals with a high electrical conductivity, where the electron mean free path between collisions is large, there exists also a possibility of observing cyclotron resonances in an ESR experiment due to the magnetic-force-induced electron orbital circulation. Cyclotron resonances are generally not easy to distinguish from true ESR resonances. In QC's the appearance of cyclotron resonances is very unlikely due to the high resistivity of the material, which implies a rather short mean free path that quenches the electron cyclotron motion in a magnetic field.

III. ESR IN MAGNETIC QUASICRYSTALS

While some QC families, like the icosahedral Al-Pd-Re, are perfectly diamagnetic, others may contain localized paramagnetic centers in addition to the itinerant moments of the conduction electrons. Examples are transition-metal-containing icosahedral families Al-Pd-Mn and Al-Cu-Fe, where Mn and Fe atoms possess unpaired *d* electrons. In such a case two kinds of electronic resonances may be observed—that of the conduction electrons and that of the localized electronic spins. As discussed above, the conduction-electron resonance is expected close to $g_F \approx 2$. The resonances of the iron group atoms are, however, also expected close to the free-electron resonance, so that the frequencies of the two systems—the conduction electrons and the localized *d* moments—should be very close. For the iron group atoms the CEF perturbing Hamiltonian is usually much larger that the spin-orbit coupling, which should result in small *g* shifts of the localized electron resonances.[12] There is an important difference between the spin-orbit-coupling-induced *g* shifts of the conduction electrons and of the localized moments. A localized electron is moving in a spherical electrostatic potential of its atom and the effect of the CEF is to quench the electronic orbital angular momentum, whereas this does not affect the atomic potential itself greatly. The resulting *g* shifts are therefore small. A conduction electron, on the other hand, is moving in the total potential of the crystal. For regular metals this potential has the periodicity of the lattice. In QC's the lattice periodicity is lacking, so that there exist significant local variations of the potential with no translational periodicity. This introduces a distribution of *g* shifts that are not necessarily small.

The *s* conduction electrons and the *d* localized spins are coupled by an exchange interaction of the form

$$\mathcal{H}_{\rm ex} = \sum_i \int d^3\vec{R} J_{sd}(\vec{R}_i - \vec{R}) \vec{S}_{di}(\vec{R}_i) \cdot \vec{S}_F(\vec{R}_i - \vec{R}). \quad (1)$$

Here J_{sd} is the exchange coupling constant, $\vec{S}_F(\vec{r})$ is the conduction- (free-) electron density at a point $\vec{r}$, $\vec{S}_{di}(\vec{R}_i)$ is the total *d*-electron spin of the transition-metal atom located at the lattice point $\vec{R}_i$, and the summation is over all magnetic atoms. This interaction introduces a broadening and a shift of the resonance. However, as the two resonance frequencies are nearly equal, the resonances of the conduction electrons and of the localized spins are excited simultaneously and one has to find the eigenfrequencies of the coupled spins.[13] The resonance frequency becomes a function of the frequencies of the two subsystems, the exchange parameter J_{sd} and the damping (the spin-lattice relaxation) of the two systems. The situation is somewhat simplified in the case when the relaxation of conduction electrons is very short,[14] as then the average electronic field may be defined (proportional to $\chi_P J_{sd}$) that shifts the *d*-spin resonance in analogy to the Knight shift of the nuclear resonance. The exchange coupling also produces a strong relaxation of both the localized and the conduction electrons. For localized spins, the relaxation rate may be calculated exactly and varies with temperature as[15] $T_1^{-1} \propto k_B T J_{sd}^2$. This rate is usually very large because J_{sd} is large, leading to a large broadening of the resonance line varying as $k_B T$ in the high-temperature range.

IV. EXPERIMENTAL TECHNIQUES AND RESULTS

In ESR spectroscopy two types of experiments are commonly employed. In the majority of cases one applies the reflection technique, where the sample is placed at the end of a resonating cavity. The measured quantity is the variation of the absorbed power by the sample when the external field is varied. In our case the reflection experiment was performed with an *X*-band instrument (irradiation frequency at 9.6 GHz, corresponding to the center absorption field of 3400 G for $g_F = 2$) on an icosahedral $Al_{63}Cu_{25}Fe_{12}$ powdered sample. A magnetic field sweep of 0–1 T was used. Magnetic susceptibility measurements[16] on several Al-Cu-Fe samples with composition nominally identical to ours (of resistivity about $\rho_{300\,\rm K} = 2600\ \mu\Omega$ cm) estimated a fraction 10^{-4} of the total number of Fe atoms to carry magnetic moments with spin $S = 2$. In a temperature run from room temperature to liquid He, the ESR signal could be detected only below 60 K, where a relatively narrow line of width $\Delta H_{pp} = 4.5–5$ G appeared [Fig. 1(a)]. Its *g* factor varied only insignificantly down to 4 K in the range $g = 2.0060–2.0063$, whereas the line intensity increased upon cooling in a Curie $1/T$ manner [Fig. 1(b)]. The signal was confirmed to originate from the QC sample by removing it from the resonator. It is, however, difficult to attribute the observed relatively narrow ESR signal to the genuine paramagnetic moments of the Al-Cu-Fe quasicrystal. It is more likely that it originates from impurities that are within the sample, but otherwise extrinsic to the quasicrystalline structure.

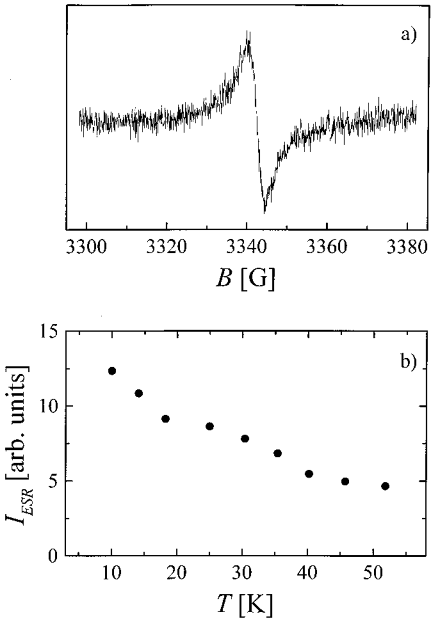

FIG. 1. (a) ESR signal of the icosahedral $Al_{63}Cu_{25}Fe_{12}$ quasicrystal at 25 K, obtained by a standard *X*-band reflection ESR spectrometer ($\nu = 9.6$ GHz). (b) The intensity of the ESR signal (corresponding to the static ESR susceptibility) as a function of the temperature.

For metallic samples, another ESR technique—the transmission technique[17,18]—has an important advantage over the reflection technique. This method takes advantage of the fact that in metals two very different characteristic lengths exist. First is the skin depth δ that determines the penetration of the microwave field into the sample. Second is the diffusion length of the electronic transverse magnetization during its lifetime $L = \sqrt{DT_1}$, where *D* is the electronic diffusion constant. One typically finds $L \gg \delta$, so that the transverse magnetization excited within the surface layer spreads with time

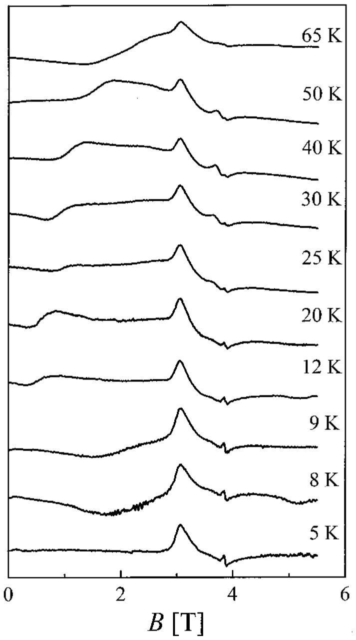

FIG. 2. Temperature-dependent ESR signals of the $Al_{63}Cu_{25}Fe_{12}$ quasicrystal, obtained by a high-field transmission ESR spectrometer ($\nu = 109.270$ GHz). The signals appear in the mixed dispersion-absorption mode.

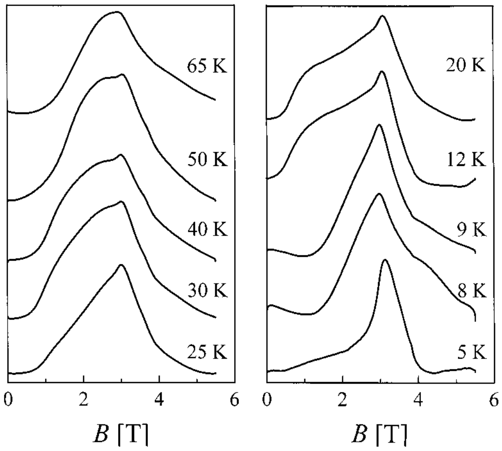

FIG. 3. High-field ESR absorption spectra of the $Al_{63}Cu_{25}Fe_{12}$ quasicrystal, obtained by phase-correcting the spectra of Fig. 2 into a pure dispersion mode, followed by an integration.

deeply into the bulk. In an ideal transmission ESR experiment one uses a bulk metallic sample of thickness l that is large compared to δ but small compared to L. The microwave frequency is applied at one of the faces of the sample and the transmitted transverse magnetization is measured at the other side. Here only the signal transferred through the sample by the conduction electrons due to their diffusion motion is observed, whereas the signals of the localized moments as well as spurious signals due to impurities in the cavity are not detected. For powdered samples with fine grains, the signals of the localized moments are detected too.

The ESR experiment on the same $Al_{63}Cu_{25}Fe_{12}$ sample was repeated on a high-field transmission ESR spectrometer, operating at the microwave frequency 109.270 GHz (corresponding to the center absorption field of $B = 3.890$ T for $g_F = 2$). The field was swept in the range 0–6 T. The spectrometer is designed to operate in fields up to 14 T and is almost identical to that described by Muller *et al.*,[19] except that it uses a Gunn diode as the radiation source. There is no resonant cavity in this case, but the transmitted signal is detected by a cooled bolometer. In this high-field transmission experiment the ESR signal of the investigated $Al_{63}Cu_{25}Fe_{12}$ quasicrystal was clearly detected. The measurements were performed at temperatures below 65 K. The signal is very broad, extending typically over a several (up to 5) T field range. A collection of high-field raw ESR spectra is displayed in Fig. 2. The spectra in this experiment do not appear in a pure dispersion mode, but exhibit a slight admixture of the absorption. A weak narrow signal is observed close to the free-electron position at $B = 3.868$ T, which is obviously the same as that observed in the X-band experiment (Fig. 1). The signals were phase corrected into pure dispersion and then integrated in order to obtain pure absorption line shapes. The absorption spectra are displayed in Fig. 3 and we discuss in the following the ESR results of the $Al_{63}Cu_{25}Fe_{12}$ quasicrystal on their basis.

V. DISCUSSION

The several-tesla broad ESR absorption spectra of Fig. 3 exhibit a not-too-well pronounced structure of two lines. The peak of the spectrum is centered at 3.1 T and its position does not shift noticeably with temperature. At present it is difficult to analyze the structure of the spectrum more than just qualitatively. It is easy to anticipate that the spectrum should show a structure of two lines, one emerging from the conduction electrons and the other from the localized Fe d moments. However, as we mentioned before, the two subsystems cannot be treated independently, but the total Hamiltonian should be diagonalized in order to obtain the position and the shape of the spectrum. With the lack of a satisfactory quasicrystalline Hamiltonian this seems to be beyond the possibilities at present.

We can now understand why the X-band experiment was unsuccessful to record the ESR spectrum of the quasicrystalline $Al_{63}Cu_{25}Fe_{12}$. The magnetic field sweep range of a conventional X-band instrument extends over the interval 0–1 T, which is much too narrow to detect the full quasicrystalline signal of several tesla width. The narrow line displayed in Fig. 1 thus represents only a tiny portion of the total ESR signal in the vicinity of $g_F = 2$. The same narrow line is also consistently observed in the high-field experiment. As discussed before, this line seems to originate from impurities extrinsic to the QC.

The width of the ESR spectrum exhibits interesting temperature dependence (Fig. 4). From 65 to about 20 K the spectrum broadens continuously upon cooling. Its full width at half height (FWHH) $\Delta H_{1/2}$ amounts to 2 T at 65 K and increases to 3 T at 20 K. Below 20 K an anomalous narrowing of the spectrum is observed, where the line narrows to 0.8 T at 5 K, thus by a factor 3.7. Since we have no model of the ESR line shape as yet, we can discuss this behavior only

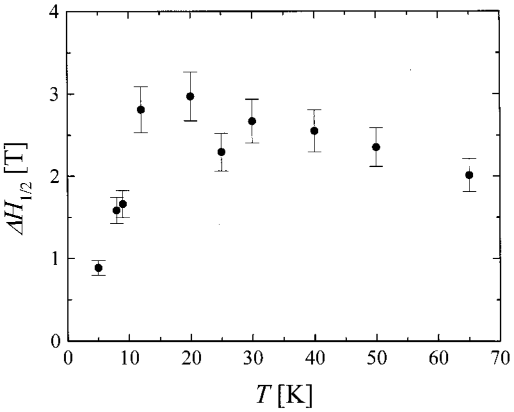

FIG. 4. Temperature dependence of the FWHH of the high-field ESR absorption spectra from Fig. 3.

qualitatively by noting that a similar anomalous temperature dependence of the spectral width was observed recently by ^{27}Al NMR in icosahedral $Al_{72.4}Pd_{20.5}Mn_{7.1}$ (Ref. 4) and by ^{27}Al and ^{55}Mn NMR in $Al_{72}Pd_{19.5}Mn_{8.5}$ (Ref. 20). Upon cooling the ^{27}Al and ^{55}Mn NMR spectra showed a $1/T$ broadening in the high-temperature regime above about 20 K and a considerable line narrowing for factors 2–10 below. This anomalous linewidth behavior was analyzed in terms of the conduction-electron screening of the manganese *d* moments within the aluminum-rich environment that takes place at low temperatures. The screening results in a reduction of the strength of the *s-d* exchange interaction that represents one of the line broadening mechanisms in NMR. The existence of the screening in the icosahedral Al-Pd-Mn QC's was recently demonstrated also by magnetic susceptibility measurements.[21] For icosahedral Al-Cu-Fe the same effect was not reported as yet, but in view of the similarity to the icosahedral Al-Pd-Mn it should not be too surprising to observe it also in this compound. In the case of $Al_{63}Cu_{25}Fe_{12}$, the localized magnetic moments are those of the Fe atoms and, since only 10^{-4} of all Fe atoms possess moments, the moments are diluted. The resistivities of the icosahedral Al-Cu-Fe and the icosahedral Al-Pd-Mn are about the same, so that a similar concentration of conduction electrons should exist in both QC's. In addition, both QC families are structurally almost identical. These arguments indicate that the screening of the localized Fe *d* moments may be responsible for the anomalous temperature dependence of the ESR linewidth also in $Al_{63}Cu_{25}Fe_{12}$. The screening would drastically reduce the strength of the *s-d* exchange interaction below a certain temperature and result in a narrowing of the ESR line.

ESR has an important advantage over NMR in detecting the changes in the *s-d* exchange coupling, emerging, e.g., due to screening of the localized *d* moments. In NMR the exchange field of the *d* moments is detected by the nuclear spins indirectly via the polarization of the conduction-electron cloud, the indirect *s-d* exchange interaction involving a product of two exchange integrals, one between the *d* moments and the conduction electrons and the other between the conduction electrons and the nuclear spins. In ESR, the resonating conduction electrons are direct-exchange coupled to the *d* spins, the interaction involving only one exchange integral. In view of that, the ESR signal should be much more sensitive to the temperature-dependent changes in the *s-d* exchange interaction.

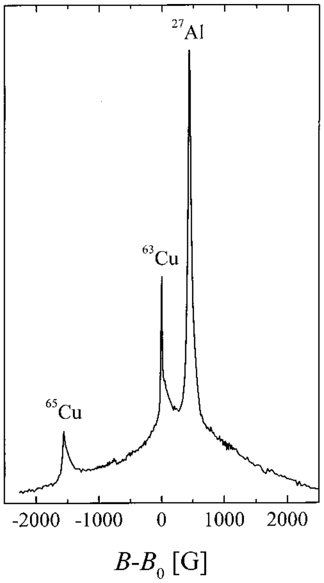

FIG. 5. NMR spectrum of the same $Al_{63}Cu_{25}Fe_{12}$ sample recorded at room temperature by a field-sweep technique at the irradiation frequency 26.134 MHz (corresponding to the center field $B_0 = 2.35$ T). Three overlapping resonances of ^{27}Al, ^{63}Cu, and ^{65}Cu nuclei are observed within the spectrum.

For comparison we show in Fig. 5 the NMR spectrum of the same $Al_{63}Cu_{25}Fe_{12}$ sample recorded by a field-sweep technique at the irradiation frequency 26.134 MHz (corresponding to the center field $B_0 = 2.35$ T). The spectrum is very broad, extending over the field interval of 5000 G. The details of the structure of the NMR spectra of Al-based QC's are given elsewhere.[2] Here we mention that the spectrum of Fig. 5 represents three overlapping resonances of ^{27}Al, ^{63}Cu, and ^{65}Cu nuclei. The resonances exhibit a large electric-quadrupole-induced inhomogeneous broadening due to the distribution of local electric fields. The sharp lines represent central transitions $(\frac{1}{2} \leftrightarrow -\frac{1}{2})$ of half-integer spins that are quadrupole-perturbed in second order, whereas the broad "background" lines represent first-order perturbed satellite transitions. The strong electric quadrupole broadening of the NMR spectrum gives a direct proof for the existence of a large distribution of the CEF in the investigated sample. The NMR and the ESR spectra thus both show consistently the large broadening due to the distribution of local electric fields that originates from the lack of periodicity in the QC lattice.

VI. CONCLUSIONS

Since ESR spectroscopy in QC's is capable of detecting signals from both the itinerant conduction electrons and the localized moments of magnetic atoms, it can provide important information on the electronic properties of quasicrystalline materials. ESR spectra of QC's are difficult to record due to the very broad signal, extending over a magnetic-field interval as large as 4–5 T. This fact makes the use of a conventional *X*-band ESR instrument that employs a relatively narrow field sweep of 0–1 T inconvenient. A high-field transmission instrument with a large field sweep of several tesla is capable of recording the full ESR spectrum of a QC sample. The origin of the strong broadening of the spectra in the investigated icosahedral $Al_{63}Cu_{25}Fe_{12}$ QC is twofold. The first is the distribution of local electric fields due to the lack of periodicity in the QC lattice and the second is the *s-d* exchange interaction between the conduction electrons and the localized Fe *d* moments. Both broadening mechanisms are consistently observed in the ESR as well as in the NMR experiments on transition-metal-containing QC's. An anomalous temperature dependence of the ESR linewidth that narrows from 20 to 5 K by a factor 3.7 may originate from the change in the strength of the *s-d* exchange interaction due to the conduction-electron screening of the localized Fe *d* moments, in analogy to the similar NMR observation in the icosahedral Al-Pd-Mn.

ACKNOWLEDGMENTS

The financial support offered by the CNRS in France and the Ministry of Education, Science and Sport in Slovenia under bilateral Project No. 8705 is gratefully acknowledged.

---

[1] A. Shastri, F. Borsa, D. R. Torgeson, J. E. Shield, and A. I. Goldman, Phys. Rev. B **50**, 15 651 (1994).

[2] T. Apih, M. Klanjšek, D. Rau, and J. Dolinšek, Phys. Rev. B **61**, 11 213 (2000).

[3] T. Apih, O. Plyushch, M. Klanjšek, and J. Dolinšek, Phys. Rev. B **60**, 14 695 (1999).

[4] J. Dolinšek, M. Klanjšek, T. Apih, J. L. Gavilano, K. Giannò, H. R. Ott, J. M. Dubois, and K. Urban, Phys. Rev. B **64**, 024203 (2001).

[5] J. Dolinšek, T. Apih, M. Simsič, and J. M. Dubois, Phys. Rev. Lett. **82**, 572 (1999).

[6] J. Dolinšek, B. Ambrosini, P. Vonlanthen, J. L. Gavilano, M. A. Chernikov, and H. R. Ott, Phys. Rev. Lett. **81**, 3671 (1998).

[7] J. Dolinšek, M. Klanjšek, T. Apih, A. Smontara, J. C. Lasjaunias, J. M. Dubois, and J. S. Poon, Phys. Rev. B **62**, 8862 (2000).

[8] F. S. Pierce, S. J. Poon, and Q. Guo, Science **261**, 737 (1993).

[9] S. K. Misra, L. E. Misiak, D. Bahadur, V. Srinivas, and R. A. Dunlap, Phys. Rev. B **40**, 7537 (1989).

[10] D. Bahadur, C. M. Srivastava, M. H. Yewondwossen, and R. A. Dunlap, J. Phys.: Condens. Matter **7**, 9883 (1995).

[11] J. Winter, *Magnetic Resonance in Metals* (Clarendon, Oxford, 1971), p. 162.

[12] See, e.g., C. P. Slichter, *Principles of Magnetic Resonance*, 2nd ed. (Springer, Berlin, 1980), p. 296.

[13] M. Peter, J. Dupraz, and H. Cottet, Helv. Phys. Acta **40**, 301 (1967).

[14] H. Hasegawa, Prog. Theor. Phys. **21**, 483 (1959).

[15] J. Winter, *Magnetic Resonance in Metals* (Ref. 11), p. 182.

[16] J. C. Lasjaunias, Y. Calvayrac, and Hongshun Yang, J. Phys. I **7**, 959 (1997).

[17] S. Vanderven and T. R. Schumacher, Phys. Rev. Lett. **12**, 695 (1964).

[18] R. B. Lewis and T. R. Carver, Phys. Rev. Lett. **12**, 693 (1964).

[19] F. Muller, M. A. Hopkins, N. Coron, M. Grynberg, L. C. Brunel, and G. Martinez, Rev. Sci. Instrum. **60**, 3681 (1989).

[20] J. Dolinšek, P. Jeglič, M. Klanjšek, T. Apih, A. Bilušić, and A. Smontara (unpublished).

[21] V. Simonet, F. Hippert, M. Audier, and G. Trambly de Laissardière, Phys. Rev. B **58**, R8865 (1998).